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00041 Nanomaterials for water treatment

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Abstract

The pressing need for advanced yet cost-effective water treatment has led to the development of a wide range of engineered nanomaterials (ENMs), from carbon-based to metal and metal oxide nanomaterials, and including novel structures such as metal oxide frameworks (MOFs) and covalent organic frameworks (COFs). ENMs have been developed to treat conventional water pollutants as well as emerging contaminants, and address new challenges. In this article we provide an overview of the current literature on a wide range of ENMs as well as several major classes of water contaminants.

Key Points

At the end of this article, readers should understand that:

- Nanomaterials can effectively treat many legacy and emerging water contaminants.
- Adsorption and/or degradation of contaminants are the main treatment mechanisms.
- Novel nanohybrids and nanocomposites can be tailored to enhance specificity and removal of contaminants.
- The largest barrier to general commercialization is cost of nanomaterials vs. conventional technology.

Introduction

Water is one of the most important resources for the survival and health of human and other organisms. However, according to a World Health Organization (WHO) report, about 29% (2.2 billion) of the world population lacked drinking water with adequate sanitation in 2017 (**World Health Organization, 2019**). The demand for clean water is rising with global population growth and increasing effects of environmental issues, such as drought, increasing temperatures, changing precipitation patterns, and water pollution (**Dos Santos et al., 2017; Mekonnen and Hoekstra, 2016; Vörösmarty et al., 2000**). Ecological stress is also increasing, as we divert more water for human consumption and impact water quality. Therefore, innovative water treatment is a necessity to address the scarcity of water resources and improve water quality (Mekonnen and Hoekstra, 2016; Kallis, 2010).

In wastewater treatment plants (WWTPs), most suspended solids and biodegradable organic matter can be removed by conventional technologies such as clarification and biological technologies (Sonune and Ghate, 2004; Daud *et al.*, 2015; Sustarsic, 2009). However, WWTPs face several challenges, which threaten the possibility of direct and even indirect water reuse. First, there are emerging contaminants (such as pharmaceuticals and personal care products [PPCPs], flame retardants, and per- and polyfluoroalkyl substances [PFAS]) that cannot be effectively removed by traditional wastewater treatment technologies, (Radjenović *et al.*, 2009; Radjenovic *et al.*, 2007; Kim *et al.*, 2020; Houtz *et al.*, 2018) and the relevant health-based regulations are becoming more stringent (Zuo *et al.*, 2014; Roberson, 2011). Second, a mixed waste stream from industrial, commercial, and residential activities increases the complexity of wastewater composition and the difficulty of wastewater treatment (Nödler and Scheurer, 2019; Xu *et al.*, 2019). Additionally, remote and developing regions where decentralized wastewater treatment is commonly applied have increasingly higher requirements for affordability and reliability of treatment technologies (Boller, 1997; Qu *et al.*, 2019). To achieve higher water quality from WWTPs, the effluent is usually purified with further treatment technologies, such as advanced oxidation process (AOP), (Monteoliva-García *et al.*, 2020; Qin *et al.*, 2015; Ahmed *et al.*, 2020; Banks *et al.*, 2020; Nzeribe *et al.*, 2019) and reverse osmosis (RO), (Mastropietro *et al.*, 2021; Abejón *et al.*, 2015; Sahebi *et al.*, 2020). However, these technologies have high operation and maintenance costs, high energy consumption, and poor selectivity, making them difficult to promote in smaller communities and developing countries (Ahmed *et al.*, 2020; Mastropietro *et al.*, 2021; Abejón *et al.*, 2015; Brienza and Katsoyiannis, 2017; Kanakaraju *et al.*, 2018).

Nanomaterials, with a size of 1–100 nm in at least one dimension, (**Ikhmayies, 2014**) have been applied in laboratory- or pilot-scale studies for environmental remediation (**Adeleye et al., 2016a**). The extremely small size of nanomaterials results in unique properties compared to larger-sized materials with a similar composition (Lin *et al.*, 2008; Rahmani *et al.*, 2011; Stanley-Wood and Lines, 2007). Nanomaterials have a much larger surface area that can contact more contaminants, leading to a higher adsorption or degradation capacity (Shaji and Zachariah, 2017; Virkutyte and Varma, 2014). Also, the increase of binding sites on the nanomaterial surface results in higher reactivity and, in some cases, can be used for water disinfection (Jiang *et al.*, 2008; Huo *et al.*, 2020; Zeng *et al.*, 2017). Thus, with high surface area and reactivity, nanomaterials are considered a prospective technology to improve water quality (**Adeleye et al., 2016a**).

This article will introduce several different types of nanomaterials, as well as their latest applications in water treatment, including the removal of different types of pollutants and the mechanisms of remediation. Additionally, this article will discuss the outlook of

nanomaterials in water remediation, in addition to discussing a few case studies that capture the application of nanotechnology in water treatment.

Classification of Nanomaterials Used for Water Treatment

Carbonaceous Nanomaterials

Carbonaceous engineered nanomaterials (C-ENMs) are mainly composed of carbon atoms. Common C-ENMs include carbon nanotubes (CNTs), (Wang *et al.*, 2019; Kumar *et al.*, 2014; Onyancha *et al.*, 2021) graphene and graphene-based materials, (Wang *et al.*, 2013; Pan *et al.*, 2020; Goodwin *et al.*, 2018) graphene oxide (GO), (Wang *et al.*, 2019; Pan *et al.*, 2020; Pendolino and Armata, 2017) fullerene, (Regulska *et al.*, 2019) and carbon nanofibers (Ding *et al.*, 2018; Painuli *et al.*, 2021). These materials have sp^2 or sp^3 (in GO or fullerene) hybrid carbon bonds and excellent physicochemical properties at the nanoscale (Goodwin *et al.*, 2018; Shan *et al.*, 2017). The large surface area and irregular morphology exhibited by C-ENMs provide a large number of binding sites for pollutants (Abd-Elhamid *et al.*, 2019). Some C-ENMs, such as fullerene, have a closed cage structure that can capture contaminants (Shan *et al.*, 2017; Kuganathan *et al.*, 2019). In addition, the wide distribution of sp^2 hybrid carbon bonds results in high electron transport, which can increase reaction rates and efficiency (Ikram *et al.*, 2020; Fan *et al.*, 2012). Further modification of C-ENMs with different functional groups can take advantage of hydrophobic or electrostatic effects to improve the selectivity of specific pollutants (Chen *et al.*, 2009; Ali *et al.*, 2019b; Ma *et al.*, 2017; Barrejón *et al.*, 2019). Thus, C-ENMs are widely considered for water purification based on mechanisms such as adsorption, (Liu *et al.*, 2020a; Zhou *et al.*, 2015; Lal *et al.*, 2020; Huang *et al.*, 2018b) filtration, (Matharu *et al.*, 2020; Cheng *et al.*, 2019b; Wang *et al.*, 2016c) and transformation (Huang *et al.*, 2018b; Yang *et al.*, 2019b). More information about the classes, mechanisms, and applications of C-ENMs have been summarized in several reviews (Shan *et al.*, 2017; Oyekunle *et al.*, 2021; Nasrollahzadeh *et al.*, 2021).

Metals and Metal Oxides

Metals and metal oxides (Me/MeO) are a class of nanomaterials composed of single or multiple metals and/or their oxides. Several nanomaterials, such as pristine and modified nanoscale zero-valent iron (nZVI), (Wang *et al.*, 2016a; Černíková *et al.*, 2020; Liu *et al.*, 2019a) nano iron oxide (Fe_2O_3), (Nogueira *et al.*, 2018; Mirbagheri and Sabbaghi, 2018; Khan and Lo, 2017) nano titanium dioxide (TiO_2), (Nogueira *et al.*, 2018; Apopei *et al.*, 2020; Chen *et al.*, 2015; de S. Furtado *et al.*, 2021; Franz *et al.*, 2020) nano zinc oxide (ZnO), (Amde *et al.*, 2020; Kamaraj *et al.*, 2020; Sun *et al.*, 2018) have been widely studied and applied for water treatment. The high remediation performance of Me/MeO not only relies on their large surface area, but also their redox properties, (Fang *et al.*, 2020; Ou *et al.*, 2020; Liu *et al.*, 2021) or the promotion of co-precipitation and complexation (Liu *et al.*, 2021; Wen *et al.*, 2014; Okonji *et al.*, 2020). Several reviews have discussed the mechanisms and applications of nano Me/MeO for water treatment (Santhosh *et al.*, 2016; Pradeep, 2009; Ighalo *et al.*, 2021; Stefaniuk *et al.*, 2016; Lee and Park, 2013).

Nanohybrids and Nanocomposites

A nanohybrid combines two or more nanomaterials, while a nanocomposite is a combination of nanomaterials and other larger-size materials such as polymeric structures or activated carbon (Aich *et al.*, 2014). Nanohybrids and nanocomposites often have the characteristics of their various constituent materials, which can make up for the shortcomings of using a single material (Mirbagheri and Sabbaghi, 2018; Nabi *et al.*, 2020; Xu *et al.*, 2018; Mehrabi *et al.*, 2019; Nawaz *et al.*, 2017). For example, doping nano TiO_2 in a polymeric membrane provides reactive sites within a stable structure and can help collect and recycle the nanoparticles after treatment (Nabi *et al.*, 2020). Modifying the ratio of different components in nanohybrids and nanocomposites may result in different properties and remediation performance (Sun *et al.*, 2018; Mehrabi *et al.*, 2019; Nawaz *et al.*, 2017). For instance, in nano TiO_2 -nano ZnO hybrid material, when nano TiO_2 is the main heterogeneous photocatalyst, its reaction activity will decrease with increasing nano ZnO . However, when nano ZnO is used as the main heterogeneous photocatalyst, the reaction activity increases with increasing nano TiO_2 (Sun *et al.*, 2018). This difference may be related to the migration direction of electrons and holes in the different materials (Sun *et al.*, 2018). More information about the classes, mechanisms, and applications of nanohybrid/nanocomposite for water treatment was covered in several reviews (Kumar *et al.*, 2021; Miculescu *et al.*, 2016; Mardikar *et al.*, 2021).

Organic Framework Nanostructures

In addition to considering the use of different chemicals, the performance of nanomaterials can also be improved by developing new nanostructures. At present, the emerging new nanostructured materials in the field of water treatment include covalent organic frameworks (COFs) and metal-organic frameworks (MOFs). MOFs are porous crystalline materials with an ordered honeycomb structure formed by combining metal elements as nodes and organic linkers (Goetjen *et al.*, 2020). COFs are similar to MOFs, except that in COFs the metal nodes are replaced with non-metals (Xia *et al.*, 2021). COFs and MOFs have high surface areas, and their inherent nano-pores can be used as channels for molecular transmission. In addition, they possess low mass and good heat resistance, which make them a popular choice for novel adsorption or filtration materials (Xia *et al.*, 2021; Mon *et al.*, 2018; Rego *et al.*, 2021; Yuan *et al.*, 2019). Some review papers also summarized the properties, mechanisms, and applications of COFs/MOFs-based nanomaterials for water treatment (Xia *et al.*, 2021; Yuan *et al.*, 2019; Li *et al.*, 2019e).

Water Treatment Mechanisms

Adsorption

In terms of water treatment, adsorption is a technology that transfers and concentrates the contaminants of interest from the water phase to the surface of a solid (adsorbent). Due to the simplicity of operation, low cost, and high efficiency, adsorption is a widely preferred method for water treatment. Many C-ENMs, (Sun *et al.*, 2020; Ahsan *et al.*, 2019; Vajedi and Dehghani, 2019; Henriques *et al.*, 2016; Ma *et al.*, 2018) nano Me/MeO, (Vajedi and Dehghani, 2019; ElHusseini *et al.*, 2020; Clark *et al.*, 2019; Irshad *et al.*, 2019; Behjati *et al.*, 2018; Rahdar *et al.*, 2021) and new generation nanostructured materials (such as COFs and MOFs (Ahsan *et al.*, 2019; Liu *et al.*, 2018; Ji *et al.*, 2018; Liu *et al.*, 2020b; Cui *et al.*, 2020a)) can adsorb contaminants, at times with particular specificity, depending on the adsorbent's chemical composition and surface characteristics.

C-ENMs, such as CNTs, graphene, and graphene-based nanomaterials, or the new generation of nanostructured materials (COFs or MOFs), have extremely high surface area and porosity, which promote interactions between the adsorbents and contaminants. They are particularly suited to remove a wide range of organic molecules, (Akinpelu *et al.*, 2019; Ahlawat *et al.*, 2020; Hashem *et al.*, 2019; Zebardast *et al.*, 2018; Wang *et al.*, 2010) but can also be tailored to remove inorganic ions (Vajedi and Dehghani, 2019; Liu *et al.*, 2020b; Samuel *et al.*, 2019; Niu *et al.*, 2021; Cui *et al.*, 2019). The adsorption mechanisms include covalent bonds, ion exchange, π -electron coupling, hydrogen bonding, electrostatic interaction, and micropore filling (Ahlawat *et al.*, 2020; Gupta *et al.*, 2013; Jabbari *et al.*, 2016). Further modification of nanomaterials with functional groups can enhance the adsorption capacity or contaminant selectivity by changing the properties of the adsorbents, such as hydrophobicity (Ahlawat *et al.*, 2020; Yang *et al.*, 2015; Norzilah *et al.*, 2011).

Me/MeO ENMs, such as nZVI, (Fang *et al.*, 2020; Han *et al.*, 2021; Wei *et al.*, 2018) nanoFe₃O₄, (Mahdavi *et al.*, 2014; Liu *et al.*, 2019c; Ali *et al.*, 2019a; Zhang *et al.*, 2019b) nanoTiO₂, (Vajedi and Dehghani, 2019; Irshad *et al.*, 2019) nanoZnO, (Mahdavi *et al.*, 2014; Gu *et al.*, 2020; Sharma *et al.*, 2019) are also widely used to adsorb many heavy metals, metalloids, and inorganic salts. The high adsorption capacity of Me/MeO ENMs is not only due to their large surface area, but also metallic elements or surface functional groups that enhance the interactions between the adsorbents and contaminants, such as metal displacement reactions, (Liang *et al.*, 2021; Liang *et al.*, 2020b; Lv *et al.*, 2019) co-precipitation, (Su *et al.*, 2015; Wen *et al.*, 2014; Lv *et al.*, 2019) and complexation (Liu *et al.*, 2021; Liang *et al.*, 2021). For instance, the adsorption capacity of nZVI for phosphate was enhanced by complexation between iron and phosphate (Liu *et al.*, 2021). Also, the sulfidation of nZVI to produce sulfide-modified nZVI (SnZVI) made the surface of the nanoparticle irregular (which increased the specific surface area) and introduced sulfhydryl (-SH) groups (which co-precipitate metals), both of which increased the nanoparticle's cadmium (Cd) adsorption capacity (Su *et al.*, 2015).

Nanocomposites, such as magnetic nanoparticles coated with a porous shell that can host ligands or surfactant micelles, have also been employed successfully to remove a wide range of contaminants, such as legacy organic pollutants, (Wang *et al.*, 2009; Clark and Keller, 2012a) emerging contaminants, (Huang and Keller, 2013; Huang *et al.*, 2016a) metals, (Huang and Keller, 2015; Huang *et al.*, 2016b; Huang and Keller, 2016) natural organic matter (NOM), (Wang *et al.*, 2011) and inorganic ions (Clark and Keller, 2012b). These interactions involve covalent bonds, electrostatic interactions, and other mechanisms to adsorb the target contaminants onto the outer shell. The magnetic nanoparticles can then be removed from the treated water using a simple magnetic field, with no pressure differential or clogging. Regeneration of the ligand or surfactant micelles is done off-line, and the functionalized magnetic nanoparticles are then reused for many cycles, reducing overall cost, and recovering the valuable nanomaterial.

When applying nanomaterials, many factors that affect the adsorption efficiency need to be considered, such as ionic strength, natural organic matter (NOM), pH, temperature, stirring speed (or flow speed), water-solubility, hydrophobicity and electrical properties of the target pollutants, and the isoelectric point of the nanomaterials (Ahsan *et al.*, 2019; Clark *et al.*, 2019; Norzilah *et al.*, 2011; Samuel *et al.*, 2018). Some metal-based nanomaterials, such as nZVI, can dissolve under acidic (low pH) conditions and reduce their effectiveness in water (Bae and Hanna, 2015). Also, a change of pH can result in different electrical properties of the adsorbate and the adsorbent, and affect the interactions occurring by electrostatic force (Ahsan *et al.*, 2019; Clark *et al.*, 2019; Norzilah *et al.*, 2011). Therefore, finding suitable reaction conditions can improve adsorption efficiency. Given the finite number of sorption sites, the nanomaterial (adsorbent) needs to be regenerated or replaced on a regular interval (Tan *et al.*, 2015; Aigbe and Osibote, 2020; Huang *et al.*, 2018a).

Filtration

Filtration is a technology that uses porous materials, including membranes, to separate chemical or biological contaminants from water under pressure. Some nanostructures that have nanoscale pores can be directly applied for filtration (Apopei *et al.*, 2020; Chen *et al.*, 2020a; Yanez *et al.*, 2017). Many nanomaterials, such as nanofibers, can be layered to generate a mesh for filtration (Chen *et al.*, 2020b; Feng *et al.*, 2013; Nalbandian *et al.*, 2015). Nanofibers have been widely used in the nanofiltration (NF) step of water treatment plants and have proven to effectively remove contaminants that are equal or higher than the molecular weight cut-off (MWCO) (Zhang *et al.*, 2019e; Ma *et al.*, 2014; Han *et al.*, 2020b). The main mechanisms of nanomaterials for filtration are size exclusion, hydrophobic interactions, and electrostatic interactions (Ganiyu *et al.*, 2015; Dolar *et al.*, 2013; Nghiem *et al.*, 2006; Alturki *et al.*, 2010). Size exclusion is often the most important filtration mechanism, (Auset and Keller, 2006; Sirivithayapakorn and Keller, 2003) and filtration capacity is often affected by the pore size and distribution of the material. Hydrophobic interactions also influence adsorption phenomena between contaminants and the membrane surface (Dolar *et al.*, 2013; Alturki *et al.*, 2010). Hydrophobic contaminants are more likely to adsorb to a C-ENM membrane surface. However, diffusion may also occur, and the adsorbed contaminant could desorb into the permeate (Dolar *et al.*, 2013; Alturki *et al.*, 2010). This phenomenon was observed when removing hydrophobic pharmaceuticals ($\log K_{ow} > 2.0$), such as procaine ($\log K_{ow} = 2.14$) (Dolar *et al.*, 2013). Electrostatic interactions also play an important role in nanofiltration (Nghiem *et al.*, 2006; Ozaki *et al.*,

2008; Verliefde *et al.*, 2007). For example, ibuprofen and diclofenac (which are both negatively charged at pH 7) have higher removal than atenolol (which is positively charged at pH 7) if the NF membrane has a positive charge at pH 7 (Verliefde *et al.*, 2007). Similarly, sulfamethoxazole was observed to possess the same charge (negative) as the nanofiltration membrane when pH was higher than its pK_a (pH 5.7), which resulted in a higher rejection rate (Nghiem *et al.*, 2006).

Many factors need to be considered in the application of NF, such as the size and hydrophobicity of the target pollutants, the pore size of the nanomaterials, the hydrophilicity and the functional groups contained, and the characteristics of the water (such as pH, ionic strength, NOM concentration, temperature) (Ganiyu *et al.*, 2015; Dolar *et al.*, 2013; Nghiem *et al.*, 2006; Alturki *et al.*, 2010). In addition, after a certain operation time, the filtration sites of porous materials and membranes become saturated, increasing the pressure differential across the membrane, and reducing water flow (Xu *et al.*, 2019; Nghiem and Hawkes, 2009). Given the nanoscale pore size and high operating pressure, the quality requirement of the influent water should be high enough to avoid fouling and blocking. Thus, before nanofiltration, the influent is often pretreated with adsorption and/or microfiltration to remove larger particles and improve the service life and energy efficiency of the NF membranes (Xu *et al.*, 2015; Kim *et al.*, 2011; Davood Abadi Farahani *et al.*, 2016).

Oxidation-Reduction

Applying redox reactions in water treatment has attracted widespread attention because of fundamental transformation or elimination of the target contaminants. The extremely high specific surface area of nanomaterials, arising from small size, leads to high reactivity and reaction kinetics. Nanomaterials may play different roles in redox reactions (such as reducing agents, (Meng *et al.*, 2021; Xu *et al.*, 2020; Lin *et al.*, 2018; Wang *et al.*, 2020b) oxidizing agents, (Babuponnusami and Muthukumar, 2012; Zha *et al.*, 2014; Mikhailov *et al.*, 2017) precursors, (Wang *et al.*, 2020e; Angamuthu *et al.*, 2017) or catalysts, (de S. Furtado *et al.*, 2021; Franz *et al.*, 2020; Ran *et al.*, 2020; Pirsaeheb *et al.*, 2020)) in the treatment of pollutants. Some nanomaterials may also transform contaminants by producing free radicals with strong oxidizing properties (indirect oxidation). For example, the dissolved iron from nZVI can lead to Fenton-like reactions in water and generate hydroxyl radical that has a strong oxidation potential (Babuponnusami and Muthukumar, 2012; Zha *et al.*, 2014; Mikhailov *et al.*, 2017). By modifying nanomaterials to improve their electron transport efficiency, their redox ability can be further improved (Lin *et al.*, 2017; Nguyen *et al.*, 2021; Deng *et al.*, 2018; Su *et al.*, 2018a). Nanomaterials such as nZVI exhibit reducing properties which may be employed for water treatment (Černíková *et al.*, 2020; Liu *et al.*, 2019a; Han *et al.*, 2021; Němeček *et al.*, 2016). Reduction can be used to immobilize heavy metals (e.g., lead [Pb], chromium [Cr]) and metalloids (e.g., arsenic [As]), (Černíková *et al.*, 2020; Liu *et al.*, 2019a; Han *et al.*, 2021; Němeček *et al.*, 2016; Su *et al.*, 2014a; Stevenson *et al.*, 2017) or for dehalogenation of solvents (e.g., tetrachloroethylene, trichloroethylene) (Meng *et al.*, 2021; Xu *et al.*, 2020; Lin *et al.*, 2018; Wang *et al.*, 2020b; Cao *et al.*, 2017).

In addition, nanomaterials can also act as photocatalysts in redox reactions. NanoTiO₂, nanoZnO, and to an extent, GO, are commonly used nanomaterials in water treatment-based photocatalysis. They generate hydroxyl radicals in water under light excitation, and have been used to effectively degrade several organic compounds in water (de S. Furtado *et al.*, 2021; Franz *et al.*, 2020; Ran *et al.*, 2020; Pirsaeheb *et al.*, 2020; Adeleye *et al.*, 2018). Although ultraviolet (UV) light has become a commonly used excitation light source because of its high energy photons, it increases operation and maintenance costs, requires a high energy to water ratio, with potentially additional environmental impacts from high energy use (e.g., generation of greenhouse gases and ozone-depleting substances) (Lee *et al.*, 2017; Bahnemann, 2004; Chong *et al.*, 2010). Therefore, recent research has focused on the development of nanomaterials with lower energy band gap to achieve photodegradation using visible light (or sunlight) (Chong *et al.*, 2010; Cui *et al.*, 2020b; Yentür and Dükkancı, 2020). A recent study successfully used a MoS₂/rGO/WO₃ nanohybrid to degrade 80% of carbamazepine in 20 min under visible light, (Cui *et al.*, 2020b) making it more competitive in real applications.

Disinfection

Conventional disinfection technologies, such as chlorination, ozonation, and ultraviolet (UV) irradiation, have been successfully applied in water treatment, (Song *et al.*, 2016; Ghernaout, 2017; Ghernaout, 2020) but they have some disadvantages such as generation of disinfection by-products, low disinfection efficiency, high energy consumption, and high cost of operation and maintenance (Ghernaout, 2017; Du *et al.*, 2017; Gude, 2015; Pichel *et al.*, 2019). High adsorption capacity and reactivity make nanomaterials a potentially viable option for inactivating microorganisms. The mechanisms of inactivation could be physical, such as filtration and adsorption, (Matharu *et al.*, 2020; Kim *et al.*, 2007; Ojha, 2020) or chemical, such as oxidation, (Ojha, 2020; Khalil *et al.*, 2011; Panchal *et al.*, 2020) or compromising cell membranes (Ojha, 2020; Arshad *et al.*, 2017). For example, nanoTiO₂ and nanoZnO are photocatalytic, and can generate hydroxyl radicals and weaken the activity of microorganisms under sunlight or ultraviolet light (Ojha, 2020; Khalil *et al.*, 2011; Panchal *et al.*, 2020). Nanomaterials can also achieve disinfection by biological inhibition (Ojha, 2020; Arshad *et al.*, 2017). For instance, nano-silver (Ag) is considered for disinfection because the Ag ions released by nano-Ag can damage cell membrane and inhibit DNA replication, thereby preventing the growth of microorganisms (Ojha, 2020).

Multifunctionality

Since some nanomaterials are used for water treatment based on multiple mechanisms that may occur simultaneously. For example, reduction and adsorption may occur during the removal of hexavalent Cr (Cr (VI)) or pentavalent As (As (V)) by nZVI (Černíková *et al.*, 2020; Han *et al.*, 2021; Němeček *et al.*, 2016). Some nanohybrids or nanocomposites that include multiple materials may also show multifunctionality and better remediation performance than a single nanomaterial (Liu *et al.*, 2019a; Ahmad *et al.*, 2018; Gu *et al.*, 2018; Zhou *et al.*, 2021). For example, doping magnetic Zirconium (Zr) to MOFs can effectively remove Cr (VI) by adsorption, reduction, and chelation (the capacity is 259.1 mg/g) (Zhou *et al.*, 2021). Also, the introduction of β-FeOOH with photocatalytic activity into the

nanofiltration membrane can simultaneously improve the self-cleaning ability of the filter membrane (Lv *et al.*, 2017; Wang *et al.*, 2020c; Chen *et al.*, 2020c).

Nanomaterials in Water Treatment

The following section presents examples of the use of diverse nanomaterials presented in “**Classification of Nanomaterials Used for Water Treatment**”, for the treatment of common classes of contaminants, applying the mechanisms presented in “**Water Treatment Mechanisms**”. For ease of reference, we summarize the studies in **Table 1** (emerging contaminants), **Table 2** (inorganic contaminants), **Table 3** (organic contaminants) and **Table 4** (disinfection by-products).

Emerging Contaminants

Microplastic

Use of carbonaceous nanomaterials (C-ENMs)

C-ENMs such as CNTs and GO have a high surface area and can adsorb onto microplastics (MPs), which increases the size of heteroagglomerates formed and the potential to remove microplastics from the aqueous phase. Large C-ENMs-based structures can also act as adsorbents for microplastics. For instance, a sponge-like GO modified with chitin was recently employed to remove polystyrene (PS), with an adsorption capacity of 5.9–8.5 mg/g (Sun *et al.*, 2020). The removal efficiency reported in the study (72.4–89.8%) was higher than that of granular activated carbon (GAC) adsorption technology (56.9–60.9%) in traditional drinking water treatment plants (DWTPs) (Wang *et al.*, 2020a). Both C-ENMs and microplastics can be attracted to each other in water via hydrophobic interactions. Only a few studies have considered enhancing the removal of MPs with C-ENMs, and all of them were based on the adsorption (see **Table 1** and **Fig. 1(a)**).

Use of metals and metal oxides (Me/MeO)

Given the high electron transport efficiency of many nano-Me/MeO, researchers have applied Me/MeO for degradation of MPs (see **Fig. 1(a)** and **Table 1** for the examples), especially photodegradation with nanoTiO₂ or nanoZnO (Nabi *et al.*, 2020; Tofa *et al.*, 2019). The photocatalytic activities of nanoTiO₂ degraded PS, with a removal efficiency up to 98.4% in 12 h (Nabi *et al.*, 2020). Also, platinum (Pt)-doped nanoZnO achieved 94% removal of polyethylene (PE) within 90 min (Tofa *et al.*, 2019).

Use of nanohybrids/nanocomposites

Nanohybrids and nanocomposites usually take advantage of several mixed materials and result in higher treatment performance or simpler operation than single nanomaterials (Chen *et al.*, 2020a; ELMACI, 2020; Tang *et al.*, 2021). A nanocomposite made up of nanoFe₃O₄ and activated carbon adsorbed 100% of PS within 30 min which is a higher efficiency than that exhibited by most GAC-based adsorption (56.9%–60.9%) and coagulation technologies (44.5%–99%) (Wang *et al.*, 2020a). The adsorption capacity of Fe₃O₄-CNTs nanohybrids for PE, polyethylene terephthalate (PET), and polyamide (PA) reached 1650, 1400, and 1100 mg/g, respectively (Tang *et al.*, 2021). The Fe₃O₄ in the nanocomposite helped to recover the MP-adsorbed nanocomposite with an external magnetic field. More so, the recovered Fe₃O₄-CNTs could be recycled through thermal treatment (at 600°C) (Tang *et al.*, 2021).

Use of organic framework nanostructures

COFs and MOFs have a large number of nanopores and strong plasticity, which make them ideal materials for adsorption and filtration (Xia *et al.*, 2021; Mon *et al.*, 2018; Rego *et al.*, 2021; Yuan *et al.*, 2019). Some recent studies related to the applications of COF/MOFs for MPs removal were listed in **Table 1**. Also, adsorption is the main mechanism used to treat MPs (**Fig. 1(a)**). A recent study fabricated Zr-MOFs-based foam, which successfully adsorbed and filtered more than 80% of PS, polyvinylidene difluoride (PVDF), and poly (methyl methacrylate) (PMMA) in water (Chen *et al.*, 2020a).

Pharmaceutical and personal care products (PPCPs), and endocrine-disrupting chemicals (EDCs)

Use of carbonaceous nanomaterials (C-ENMs)

The main mechanisms for removing PPCPs and endocrine-disrupting chemicals (EDCs) with C-ENMs are adsorption and degradation (see **Fig. 1(b)**, with some examples in **Table 1**). The photoactivity of GO leads to the degradation of bisphenol A (BPA) (Adeleye *et al.*, 2018). The most commonly used sorbents are CNTs, including single-walled (SWCNTs) (Kim *et al.*, 2014; Balarak *et al.*, 2016; Xu *et al.*, 2017a) or multi-walled carbon nanotubes (MWCNTs) (Kim *et al.*, 2014; Xu *et al.*, 2017a; Wang *et al.*, 2016b). Due to the graphite layer present in CNT, they have a very high van der Waals force index (Liu *et al.*, 2013; Ji *et al.*, 2009). Also, CNTs are composed of fused benzene rings with strong polar sp² hybrid carbon atoms, which have a good affinity for aromatic or hydrophobic pollutants (Liu *et al.*, 2013; Lin and Xing, 2008; Das *et al.*, 2014). Studies have confirmed that the adsorption capacity of CNTs for BPA is as high as 82.9 mg/g (Ahsan *et al.*, 2019). Additionally, C-ENMs have abundant modifiable sites, and by adding appropriate functional groups, the adsorption capacity and the selectivity for the contaminants can be enhanced (Wang *et al.*, 2016b; Jung *et al.*, 2015; Piao *et al.*, 2008; Lee *et al.*, 2016).

Table 1 Recent studies about applying nanomaterials for emerging contaminants removal

Contaminant	Technology	Method	Mechanisms	Efficiency (time)	Capacity (mg/g)	Referen
<i>Microplastics (MPs)</i>						
Polystyrene (PS)	Me/MeO Nanohybrid	TiO ₂ + UV Zr-MOF	Photodegradation Adsorption	98.4% (12 h) 85.7%		(Nabi <i>et al.</i> 2020) (Chen <i>et al.</i> 2020)
	Nanohybrid Nanocomposite	Carbon@Fe ₃ O ₄ Chitin-GO sponge	Adsorption Adsorption	100 (30 min) 72.4%–89.8%	5.898–8.461	(ELM <i>et al.</i> 2020) (Sun <i>et al.</i> 2020)
Polyethylene (PE)	Me/MeO	TiO ₂ + UV	Photodegradation	100% (36 h)		(Nabi <i>et al.</i> 2020)
	Nanohybrid Nanohybrid	Fe ₃ O ₄ -CNT ZnO-Pt	Adsorption Photodegradation	94% (90 min)	1650	(Tang <i>et al.</i> 2020) (Tofa <i>et al.</i> 2020)
Polyethylene terephthalate (PET)	Nanohybrid	Fe ₃ O ₄ -CNT	Adsorption		1400	(Tang <i>et al.</i> 2020)
Polyamide (PA)	Nanohybrid	Fe ₃ O ₄ -CNT	Adsorption		1100	(Tang <i>et al.</i> 2020)
Polyvinylidene difluoride (PVDF)	Nanohybrid	Zr-MOF	Adsorption	90.1%		(Chen <i>et al.</i> 2020)
Poly (methyl methacrylate) (PMMA)	Nanohybrid	Zr-MOF	Adsorption	88.2%		(Chen <i>et al.</i> 2020)
<i>Pharmaceuticals</i>						
Carbamazepine	Me/MeO	TiO ₂ + UV	Photodegradation	100% (1 h)		(Ran <i>et al.</i> 2020)
	Me/MeO	TiO ₂ + UV	Photodegradation	99% (55 min)		(Franz <i>et al.</i> 2020)
	Me/MeO	UiO-66	Adsorption		16.69	(ElHussein <i>et al.</i> 2020)
	Nanohybrid	CoMgFe-layered double oxides (LDO)	Redox	100% (20 min)		(Hong <i>et al.</i> 2020)
	Nanohybrid	UiO-66/graphene	Adsorption		51.17	(ElHussein <i>et al.</i> 2020)
	Nanohybrid	MoS ₂ /rGO/ WO ₃ +visible light	Photodegradation	79.67% (20 min)		(Cui <i>et al.</i> 2020)
	Nanohybrid	Ag/AgCl/ BiVO ₄ +visible light	Photodegradation	70.6% (4 h)		(Yentü <i>et al.</i> 2020)
	Nanocomposite	Fe ₃ O ₄ -black carbon	Adsorption		33.6	(He <i>et al.</i> 2020)
	Nanocomposite	CuO/Cu ₂ O/Cu-biochar composite	Adsorption	93.02% (3.5 h)	4.9	(Liang <i>et al.</i> 2020)
	Diclofenac	Me/MeO	ZnFe ₂ O ₄	Redox	90% (2 min)	
Me/MeO		Sulfide-modified nZVI (SnZVI)	Adsorption + oxidation	73.6% (2 h)		(Su <i>et al.</i> 2020)

Table 1 (Continued)

Contaminant	Technology	Method	Mechanisms	Efficiency (time)	Capacity (mg/g)	Reference	
	Nanohybrid	Co ₃ O ₄ -CeO ₂	Photodegradation	90% (15 min)		(Xian <i>et al.</i> , 2019)	
	Nanohybrid	ZnO/SnO ₂ + UV	Photodegradation	100% (1 h)		(Novakovic <i>et al.</i> , 2020)	
	Nanohybrid	ZnO/TiO ₂ + UV	Photodegradation	100% (1 h)		(Novakovic <i>et al.</i> , 2020)	
	Nanohybrid	ZnO/In ₂ O ₃ + UV	Photodegradation	100% (1 h)		(Novakovic <i>et al.</i> , 2020)	
	Nanohybrid	GO@CoFe ₂ O ₄	Adsorption		32.4	(Tran <i>et al.</i> , 2020)	
	Nanohybrid	Fe ₃ O ₄ - COF	Adsorption		40.4	(Zhuang <i>et al.</i> , 2020)	
	Nanohybrid	MWCNTs- Fe ₃ O ₄	Redox	98.52% (82.24 min)		(Pourzamani <i>et al.</i> , 2018)	
	Nanocomposite	CuO/Cu ₃ O/Cu-biochar composite	Adsorption	88.96% (3.5 h)	4.2	(Liang <i>et al.</i> , 2020a)	
	Nanocomposite	GAC-TiO ₂ N	Filtration	80%		(Apopei <i>et al.</i> , 2020)	
<i>Endocrine-Disrupting Chemicals (EDCs)</i>							
Bisphenol A (BPA)	Carbonaceous	GO	Adsorption		99.7	(Ahsan <i>et al.</i> , 2019)	
	Carbonaceous	CNT	Adsorption		82.9	(Ahsan <i>et al.</i> , 2019)	
	Me/MeO	nZVI	Redox	96.4% (2.5 h)		(Guo <i>et al.</i> , 2020b)	
	Me/MeO	amino-functionalized magnetic Fe ₃ O ₄	Redox	87.3% (11 h)		(Sadeghzadeh <i>et al.</i> , 2020)	
	Me/MeO	CoFe ₂ O ₄	Redox	97% (40 min)		(Long <i>et al.</i> , 2021)	
	Me/MeO	Peroxymonosulfate (PMS)-modified CoS	Redox	90% (10 min)		(Ding <i>et al.</i> , 2020)	
	Nanohybrid	Cu@GO	Adsorption		182.2	(Ahsan <i>et al.</i> , 2019)	
	Nanohybrid	Cu@CNT	Adsorption		164.1	(Ahsan <i>et al.</i> , 2019)	
	Nanohybrid	Fe@Nitrogen-doped CNT-rGO	Redox		~280	(Sridhar and Park, 2020)	
	Nanohybrid	TiO _{2-x} /rGO	Photodegradation	~95% (60 min)		(Xu <i>et al.</i> , 2018)	
	Nanocomposite	Polysulfone/GO membrane	Filtration	93%		(Nasseri <i>et al.</i> , 2018)	
	Nanocomposite	kaolin-based Ag@TiO ₂	Photodegradation	93.22% (3 h)		(Shareef <i>et al.</i> , 2020)	
	Organic framework nanostructures	COF	Adsorption		61.3	(Liu <i>et al.</i> , 2018)	
	<i>Per- and polyfluoroalkyl substances (PFASs)</i>						

Table 1 (Continued)

Contaminant	Technology	Method	Mechanisms	Efficiency (time)	Capacity (mg/g)	Reference
Perfluoro-n-hexanoic acid (PFHxA)	Organic framework nanostructures	[NH ₂]-COF	Adsorption	90% (0.5 h)		(Ji <i>et al.</i> , 2018)
	Nanocomposite	Ag-activated carbon	Adsorption	81.5%–83% (24 h)		(Omo-Okoro <i>et al.</i> , 2021)
Perfluoro-n-heptanoic acid (PFHpA)	Organic framework nanostructures	[NH ₂]-COF	Adsorption	94% (0.5 h)		(Ji <i>et al.</i> , 2018)
	Nanocomposite	Ag-activated carbon	Adsorption	82.2%–83% (24 h)		(Omo-Okoro <i>et al.</i> , 2021)
Perfluoro-n-octanoic acid (PFOA)	Carbonaceous	CNT	Electrochemical oxidation	> 90% (3 h)		(Xue <i>et al.</i> , 2015)
	Me/MeO	Zr-PbO ₂	Electrocatalysis	81.8% (1.5 h)		(Xu <i>et al.</i> , 2017b)
	Me/MeO	Fe-TiO ₂ , Cu-TiO ₂	Photodegradation	91% (12 h)		(Chen <i>et al.</i> , 2015)
	Me/MeO	M-TiO ₂ (Pt, Pd, Ag)	Photodegradation	57.7%–100% (7 h)		(Li <i>et al.</i> , 2016)
	Nanohybrid	TiO ₂ -MWCNT	Photodegradation	~100% (8 h)		(Song <i>et al.</i> , 2012)
	Nanohybrid	In ₂ O ₃ -Graphene \	Photodegradation	~90% (3 h)		(Li <i>et al.</i> , 2013)
	Nanohybrid	Pb-BFeO ₃ /rGO	Redox	~90% (5 min)		(Li <i>et al.</i> , 2017)
	Nanohybrid	SiC/graphene	Photodegradation	40.5%–58.5% (8 h)		(Huang <i>et al.</i> , 2016c)
	Nanohybrid	Zr-MnFe ₂ O ₄ @rGO	Adsorption		10.1	(Elanchezhian <i>et al.</i> , 2020)
	Nanohybrid	nZVI-rGO	Redox	39% (10 min)		(Masud <i>et al.</i> , 2021)
	Nanohybrid	CeO ₂ /In ₂ O ₃	Photodegradation	> 90% (1 h)		(Jiang <i>et al.</i> , 2016)
	Nanocomposite	Phosphorene nanocomposite membrane	Filtration	99%		(Eke <i>et al.</i> , 2021)
	Nanocomposite	Ag-activated carbon	Adsorption	82.4%–83% (24 h)	321.2	(Omo-Okoro <i>et al.</i> , 2021)
	Nanocomposite	SnO ₂ -Sb/carbon aerogel	Electrochemical oxidation	91% (5 h)		(Zhao <i>et al.</i> , 2013)
	Organic framework nanostructures	[NH ₂]-COF	Adsorption	95% (0.5 h)		(Ji <i>et al.</i> , 2018)
Organic framework nanostructures	MOF	Adsorption	84.53%		(Guo <i>et al.</i> , 2020a)	

Table 1 (Continued)

Contaminant	Technology	Method	Mechanisms	Efficiency (time)	Capacity (mg/g)	Reference
Perfluoro-n-nonanoic acid (PFNA)	Organic framework nanostructures	[NH ₂]-COF	Adsorption	96% (0.5 h)		(Ji <i>et al.</i> , 2018)
	Nanocomposite	Ag-activated carbon	Adsorption	82.7%–83% (24 h)		(Omo-Okoro <i>et al.</i> , 2021)
Perfluoro-n-pentanoic acid (PFPeA)	Nanocomposite	Ag-activated carbon	Adsorption	82.3%–82.9% (24 h)		(Omo-Okoro <i>et al.</i> , 2021)
	Nanohybrid	nZVI-rGO	Redox	18% (10 min)		(Masud <i>et al.</i> , 2021)
Perfluoro-n-decanoic acid (PFDA)	Organic framework nanostructures	[NH ₂]-COF	Adsorption	97% (0.5 h)		(Ji <i>et al.</i> , 2018)
	Nanocomposite	Ag-activated carbon	Adsorption	82.8%–83% (24 h)		(Omo-Okoro <i>et al.</i> , 2021)
Perfluoro-n-undecanoic acid (PFUA)	Organic framework nanostructures	[NH ₂]-COF	Adsorption	97% (0.5 h)		(Ji <i>et al.</i> , 2018)
Perfluoro-n-dodecanoic acid (PFDoA)	Organic framework nanostructures	[NH ₂]-COF	Adsorption	97% (0.5 h)		(Ji <i>et al.</i> , 2018)
Perfluoro-n-tridecanoic acid (PRTrDA)	Organic framework nanostructures	[NH ₂]-COF	Adsorption	100% (0.5 h)		(Ji <i>et al.</i> , 2018)
Perfluoro-n-butanoic acid (PFBA)	Organic framework nanostructures	[NH ₂]-COF	Adsorption	62% (0.5 h)		(Ji <i>et al.</i> , 2018)
	Nanocomposite	Ag-activated carbon	Adsorption	81%–82.6% (24 h)		(Omo-Okoro <i>et al.</i> , 2021)
Perfluorobutane sulfonate (PFBS)	Organic framework nanostructures	[NH ₂]-COF	Adsorption	88% (0.5 h)		(Ji <i>et al.</i> , 2018)
	Nanocomposite	Ag-activated carbon	Adsorption	81.8%–83% (24 h)		(Omo-Okoro <i>et al.</i> , 2021)
Perfluorohexane sulfonate (PFHxS)	Organic framework nanostructures	[NH ₂]-COF	Adsorption	96% (0.5 h)		(Ji <i>et al.</i> , 2018)
	Nanocomposite	Ag-activated carbon	Adsorption	82.4%–83% (24 h)		(Omo-Okoro <i>et al.</i> , 2021)
Perfluorooctane sulfonate (PFOS)	Me/MeO	UiO-66	Adsorption		1.24	(Clark <i>et al.</i> , 2019)
	Me/MeO	MnO ₂ + H ₂ O ₂	Redox	~99% (15 min)		(Chang <i>et al.</i> , 2020)
	Me/MeO	TiO ₂ /UV	Photodegradation	83% (8 h)		(de S. Furtado <i>et al.</i> , 2021)

Table 1 (Continued)

Contaminant	Technology	Method	Mechanisms	Efficiency (time)	Capaci. (mg/g)
Perfluoropentane sulfonic acid (PFPeS) Perfluoro-2-propoxypropanoic acid (GenX)	Nanohybrid	Pd/nZVI	Adsorption	35% (6 d)	12.6
	Nanohybrid	Ni/nZVI	Adsorption & defluorination	94% (120 h)	
	Nanohybrid	Zr-MnFe ₂ O ₄ @rGO	Adsorption		
	Nanohybrid	nZVI-rGO	Redox	84% (10 min)	
	Nanocomposite	Ag-activated carbon	Adsorption	82.9%–83% (24 h)	
	Organic framework nanostructures	[NH ₂]-COF	Adsorption	97% (0.5 h)	
	Organic framework nanostructures	biphenyl-4,4'-dicarboxylic acid (BPDC)-modified MOF	Adsorption		
	Nanohybrid	nZVI-rGO	Redox	19% (10 min)	
	Organic framework nanostructures	[NH ₂]-COF	Adsorption	91% (0.5 h)	

Use of metal and metal oxides (Me/MeO)

Like C-ENMs, Me/MeO nanomaterials are often able to adsorb, and degrade PPCPs and EDCs through redox reactions (Fig. 1(b), with some examples in Table 1) (Franz *et al.*, 2020; Ran *et al.*, 2020; Novakovic *et al.*, 2020; Hong *et al.*, 2020b; Al-Anazi *et al.*, 2020; Xian *et al.*, 2019). nanoTiO₂ and nanoZnO are the most popular choices for photocatalytic degradation of PPCPs and EDCs (Franz *et al.*, 2020; Ran *et al.*, 2020; Novakovic *et al.*, 2020). In the presence of UV, TiO₂ removed 100% of carbamazepine (which is hard to biodegrade) within 1 h (Ran *et al.*, 2020). In addition to nanoTiO₂ and nanoZnO, there are studies considering the use of iron-containing nanomaterials, such as nanoZnFe₂O₄ and SnZVI, to degrade other persistent PPCPs such as diclofenac (Su *et al.*, 2018a; Al-Anazi *et al.*, 2020). One study found that both oxidation and adsorption occurred when SnZVI is used for diclofenac removal (Su *et al.*, 2018a).

Use of nanohybrids/nanocomposites

At present, researchers are exploring new nanohybrids or nanocomposites to improve the efficiency of PPCPs and EDCs removal by nanostructures (Apopei *et al.*, 2020; ElHusseini *et al.*, 2020; Cui *et al.*, 2020b; Liang *et al.*, 2020a; Pourzamani *et al.*, 2018; Masud *et al.*, 2020). For example, Fe₃O₄-COFs or CoFe₂O₄@GO exhibited a significant improvement in the adsorption of diclofenac (Tran *et al.*, 2020; Zhuang *et al.*, 2020). The adsorption capacity of Fe@CNT-rGO for BPA was as high as 280 mg/g (Sridhar and Park, 2020). In addition, studies found that the hybrids of different metallic nanomaterials can result in improved reactivity and photocatalytic ability by changing the crystal structure of the metals (Novakovic *et al.*, 2020; Tobaldi *et al.*, 2010; Wang *et al.*, 2005). For example, the hybridizing of nanoTiO₂ by nanoZnO improved electron transport efficiency of nanoTiO₂, which result in 100% photodegradation of diclofenac within 1 h (Novakovic *et al.*, 2020). Magnetic permanently confined micelle arrays (Mag-PCMA), which contain a magnetic core with silica shell bonded with surfactant micelles, (Wang *et al.*, 2009) showed a fast and high adsorption ability for several PPCPs, including atenolol, gemfibrozil, and sulfamethoxazole (Huang and Keller, 2013; Huang *et al.*, 2016a). The remediation performance can be optimized by adding a micelle swelling agent to increase the surface area and pore volume of Mag-PCMA (Huang *et al.*, 2016a). Other studies that applied nanohybrids or nanocomposites were listed in Table 1.

Use of organic framework nanostructures

Current research on the use of new generation nanostructure materials for PPCPs and EDCs mainly focuses on adsorption (Fig. 1(b), some examples were listed in Table 1) (Liu *et al.*, 2018; Zhuang *et al.*, 2020). A recent study showed that the adsorption capacity of COFs for BPA was up to 61.3 mg/g (Liu *et al.*, 2018). while Fe₃O₄-COF, a nanohybrid, had an adsorption capacity of 40.4 mg/g for diclofenac (Zhuang *et al.*, 2020).

Table 2 Recent studies about applying nanomaterials for inorganic contaminants removal

<i>Contaminant</i>	<i>Technology</i>	<i>Method</i>	<i>Mechanisms</i>	<i>Efficiency</i>	<i>Capacity (mg/g)</i>	<i>References</i>
<i>Metal</i>						
Cadmium	Carbonaceous	Poly (2-hydroxyethyl methacrylate) grafted graphene oxide	Adsorption	89% (30 min)	10.667	(Salawudeen <i>et al.</i> , 2020)
	Carbonaceous	Sawdust-derived cellulose nanocrystals (CNC)	Adsorption		2207	(Oyewo <i>et al.</i> , 2019)
	Carbonaceous	rGO	Adsorption		8.5	(Vajedi and Dehghani, 2019)
	Me/MeO	FeS	Adsorption & co-precipitation	98.5%		(Shahryari <i>et al.</i> , 2019)
	Me/MeO	FeS (stabilized with sodium carboxymethyl cellulose)	Adsorption & co-precipitation	93% (240 min)	497.5	(Tian <i>et al.</i> , 2020)
	Me/MeO	SnZVI	Adsorption & co-precipitation		85–120	(Su <i>et al.</i> , 2015)
	Me/MeO	TiO ₂	Adsorption		9.7	(Vajedi and Dehghani, 2019)
	Me/MeO	TiO ₂	Adsorption	89.45% (120 min)	45.1	(Irshad <i>et al.</i> , 2019)
	Nanohybrid	Poly (vinyl alcohol)/chitosan on nanofibers membranes	Adsorption		148.79	(Karim <i>et al.</i> , 2019)
	Nanohybrid	Poly(lactic acid)/nano chitosan composite fibers	Adsorption	75% (180 min)		(Thomas <i>et al.</i> , 2020)
	Nanohybrid	Chitosan/phosphorylated nanocellulose	Adsorption		232.55	(Brandes <i>et al.</i> , 2019)
	Nanohybrid	Polyvinylpyrrolidone-Fe ₃ O ₄	Adsorption		43.92	(Hong <i>et al.</i> , 2020a)
	Nanohybrid	Fe ₃ O ₄ /GO	Adsorption		52.083	(Thy <i>et al.</i> , 2019)
	Nanohybrid	TiO ₂ /rGO	Adsorption		9.5	(Vajedi and Dehghani, 2019)
	Nanohybrid	Activated carbon/ZrO ₂	Adsorption		166.7	(Sharma and Naushad, 2020)
	Nanohybrid	EDTA-functionalized Fe ₃ O ₄	Adsorption & complexation		79.4	(Huang and Keller, 2015)
	Nanocomposite	γ -cyclodextrin-MOF-based nanoporous carbon	Adsorption		140.85	(Liu <i>et al.</i> , 2019b)
Organic framework nanostructures	COF	Adsorption		116	(Liu <i>et al.</i> , 2020b)	
Chromium (VI)	Carbonaceous	carbon nano-rods (functionalized with 2, 2'-(ethylenedioxy)bis (ethylamine))	Adsorption & reduction (under natural light)	~99% (180 min)		(Anand <i>et al.</i> , 2019)
	6 Me/MeO	Boehmite (γ -AlO)	Adsorption & reduction		64.7	(Li <i>et al.</i> , 2019c)

Table 2 (Continued)

Contaminant	Technology	Method	Mechanisms	Efficiency	Capacity (mg/g)	References
Copper	Nanohybrid	Fe/Al bimetallic nanoparticle	Adsorption & reduction		1470	(Ou et al., 2020)
	Nanohybrid	Polyvinylpyrrolidone-Fe ₃ O ₄	Adsorption		25.52	(Hong et al., 2020a)
	Nanohybrid	MgFe ₂ O ₄ @multi-walled carbon nanotubes (MWCNTs)	Adsorption		175.43	(Verma and Balomajumder, 2020)
	Nanohybrid	Magnetic Zr-based MOF	Adsorption & reduction & chelation		259.1	(Zhou et al., 2021)
	Nanohybrid	nZVI@MOF	Adsorption & reduction		226.5	(Fang et al., 2020)
	Nanocomposite	Chitosan-GO	Adsorption		104.16	(Samuel et al., 2019)
	Nanocomposite	Chitosan@Ag	Adsorption	90% (300 min)	0.60	(Ekambaram et al., 2020)
	Nanocomposite	ZnO-tetrapods/activated carbon	Adsorption & reduction	97% (360 min)	1.016	(Sharma et al., 2019)
	Nanocomposite	Zn-MOF/chitosan	Adsorption		225	(Niu et al., 2021)
	Organic framework nanostructures	COF	Adsorption		384	(Cui et al., 2019)
	Carbonaceous	Carbon nanotubes (CNTs) (made by orange peel)	Adsorption	94% (75 min)	16.64	(Safari et al., 2019)
	Carbonaceous	rGO	Adsorption		6.1	(Vajedi and Dehghani, 2019)
	Me/MeO	TiO ₂	Adsorption		4.2	(Vajedi and Dehghani, 2019)
	Me/MeO	Fe ₃ O ₄	Adsorption		124.8	(Liu et al., 2019c)
	Nanohybrid	TiO ₂ /rGO	Adsorption		7.8	(Vajedi and Dehghani, 2019)
	Nanohybrid	Fe ₃ O ₄ @MOF@COF	Adsorption		37.29	(Li et al., 2020)
	Nanohybrid	Fe ₃ O ₄ /triethanolamine/GO	Adsorption		68.7	(Ren et al., 2019)
Nanocomposite	Polyetherimide based polyhedral oligomeric silsesquioxane (POSS) nanofiltration membrane	Filtration & adsorption	86%		(Bandeali et al., 2019)	
Nanocomposite	Fe ₃ O ₄ @activated carbon	Adsorption		23.61	(Shahrashoub and Bakhtiari, 2021)	
Nanocomposite	Chitosan-coated carbon nanotubes (CNTs)	Adsorption		115.48	(Dou et al., 2019)	
Organic framework nanostructures	Polydopamine-COF	Adsorption		109.2 (10 min)	(Xiao et al., 2021)	
Lead	Carbonaceous	Multi-walled carbon nanotubes (MWCNTs) functionalized by selenophosphoryl groups	Adsorption		156.25 (60 min)	(Kończyk et al., 2019)

Table 2 (Continued)

Contaminant	Technology	Method	Mechanisms	Efficiency	Capacity (mg/g)	References
	Carbonaceous	rGO	Adsorption		8.6	(Vajedi and Dehghani, 2019)
	Me/MeO	TiO ₂	Adsorption		7.4	(Vajedi and Dehghani, 2019)
	Nanohybrid	TiO ₂ /rGO	Adsorption		9.1	(Vajedi and Dehghani, 2019)
	Nanohybrid	Fe ₃ O ₄ /triethanolamine/GO	Adsorption		121.5	(Ren <i>et al.</i> , 2019)
	Nanohybrid	Polyvinylpyrrolidone-Fe ₃ O ₄	Adsorption		61.67	(Hong <i>et al.</i> , 2020a)
	Nanohybrid	EDTA-functionalized Fe ₃ O ₄	Adsorption & complexation		100.2	(Huang and Keller, 2015)
	Nanocomposite	poly (vinyl alcohol)/chitosan@Nanofibers membranes	Adsorption		266.12	(Karim <i>et al.</i> , 2019)
	Nanocomposite	nZVI/activated carbon	Adsorption & reduction & precipitation	95% (5 min)	59.35	(Liu <i>et al.</i> , 2019a)
	Nanocomposite	Polyetherimide based polyhedral oligomeric silsesquioxane (POSS) nanofiltration membrane	Filtration (adsorption)	85%		(Bandehali <i>et al.</i> , 2019)
	Nanocomposite	poly (2-acrylamido-2-methylpropanesulfonic acid) graft-Ni ₃ Si ₂ O ₅ (OH) ₄ -MWCNT	Adsorption		153.3	(Xiao and Lin, 2020)
	Nanocomposite	CeO ₂ -MoS ₂ hybrid biochar	Adsorption		263.6	(Li <i>et al.</i> , 2019a)
	Organic framework nanostructures	Sulfhydryl functionalized COF (SH-COF)	Adsorption		239	(Cao <i>et al.</i> , 2020)
	Organic framework nanostructures	Amide-based COF	Adsorption		185.7	(Li <i>et al.</i> , 2019d)
Mercury	Carbonaceous	GO foam	Adsorption	96% (1440 min)	35	(Henriques <i>et al.</i> , 2016)
	Carbonaceous	SWCNTs	Adsorption		41.66	(Alijani and Shariatnia, 2018)
	Me/MeO	Fe ₃ O ₄ modified with tetraethylenepentamine)	Adsorption		109.5	(Behjati <i>et al.</i> , 2018)
	Nanohybrid	GO-Fe ₃ O ₄	Adsorption		16.6	(Diagboya <i>et al.</i> , 2015)
	Nanohybrid	Fe ₃ O ₄ @ZrP	Adsorption		181.8	(Ahmad <i>et al.</i> , 2017)
	Nanohybrid	COF-supported Ag	Adsorption	99% (30 min)	113	(Wang <i>et al.</i> , 2020d)
	Nanohybrid	Se@ZnO-nanorods	Adsorption		1110	(Amde <i>et al.</i> , 2020)
	Nanohybrid	Magnetic single walled carbon nanotubes (SWCNTs)-CoS	Adsorption		1660	(Alijani and Shariatnia, 2018)

Table 2 (Continued)

Contaminant	Technology	Method	Mechanisms	Efficiency	Capacity (mg/g)	References
Non-metal Arsenic	Nanocomposite	Cellulose nanofibrils (CNFs) @ bio-sorbent films	Adsorption	99% (1440 min)		(Silva <i>et al.</i> , 2020)
	Nanocomposite	MoS ₂ @fiber	Adsorption		6258.7	(Fausey <i>et al.</i> , 2020)
	Organic framework nanostructures	Pyrene-based COF	Adsorption		232 (Hg ⁰), 738 (Hg ²⁺)	(Cui <i>et al.</i> , 2020a)
	Organic framework nanostructures	Sulfonated COF	Adsorption	99.8% (30 min)	98.42	(Panda <i>et al.</i> , 2020)
	Carbonaceous	SWCNTs	Adsorption		49.65	(Ma <i>et al.</i> , 2018)
	Me/MeO	Maghemite (γ -Fe ₂ O ₃ functionalized by starch)	Adsorption		8.67	(Siddiqui <i>et al.</i> , 2020)
	Nanohybrid	GO-CuFe ₂ O ₄	Adsorption		51.64 (As (III)), 124.69 (As (V))	(Wu <i>et al.</i> , 2018)
	Nanohybrid	GO-Gd ₂ O ₃	Adsorption		216.7	(Lingamdinne <i>et al.</i> , 2021)
	Nanocomposite	Fe ₃ O ₄ @activated carbon	Adsorption		6.69	(Joshi <i>et al.</i> , 2019)
	Nanocomposite	Fe ₃ O ₄ @Al ₂ O ₃ @Zn-Fe layered double hydroxides (LDH)	Adsorption		67.57	(Adlnasab <i>et al.</i> , 2019)
	Nanocomposite	Nano-CeO ₂ @ceramic filter	Filtration & adsorption	94.27%		(Yang <i>et al.</i> , 2021)
	Nanocomposite	TiO ₂ @activated carbon	Adsorption		13.38 (As (III)), 15.18 (As (V))	(Luo <i>et al.</i> , 2020)
Nitrate	Nanocomposite	Fe-Ti-Mn composite oxide	Adsorption		122.3 (As (III)), 74.4 (As (V))	(Zhang <i>et al.</i> , 2019a)
	Organic framework nanostructures	Zn-MOF-74	Adsorption		211 (As (III)), 325 (As (V))	(Yu <i>et al.</i> , 2019)
	Organic framework nanostructures	ethidium bromide modified-COF	Adsorption		53.1	(Yang <i>et al.</i> , 2020)
	Me/MeO	CuO	Adsorption		71.56	(Rahdar <i>et al.</i> , 2021)
	Me/MeO	Boehmite (γ -AlO)	Adsorption		378.5	(Li <i>et al.</i> , 2019c)
	Me/MeO	nZVI	Reduction	32.8% (30 min)		(Wei <i>et al.</i> , 2018)
	Nanocomposite	nZVI@biochar	Reduction	78.2% (30 min)		(Wei <i>et al.</i> , 2018)
	Nanocomposite	nanochitosan/clinoptilolite@ pentaethylenhexamine	Adsorption		277.77	(Yazdi <i>et al.</i> , 2019)

Table 2 (Continued)

<i>Contaminant</i>	<i>Technology</i>	<i>Method</i>	<i>Mechanisms</i>	<i>Efficiency</i>	<i>Capacity (mg/g)</i>	<i>References</i>
	Nanocomposite	Chitosan nanobiopolymer@polyethersulfone (PES) membrane	Filtration	100%		(Ghaemi <i>et al.</i>, 2018)
	Organic framework nanostructures	Fe supported MOF	Adsorption & complexation		92.59	(Pandi and Choi, 2021)

Table 3 Recent studies about applying nanomaterials for organic contaminants removal

<i>Contaminant</i>	<i>Technology</i>	<i>Method</i>	<i>Mechanisms</i>	<i>Efficiency</i>	<i>Capacity (mg/g)</i>	<i>References</i>
<i>Chlorinated compounds</i>						
Trichloroethylene (TCE)	Carbonaceous	CNT	Filtration	74%		(Yanez <i>et al.</i> , 2017)
	Carbonaceous	GO	Adsorption		137.5	(Rohaizad <i>et al.</i> , 2020)
	Me/MeO	nZVI	Redox	3.38% (420 min)		(Meng <i>et al.</i> , 2021)
	Me/MeO	nZVI-alternating current electromagnetic field (AC EMF)	Dichlorination	~100% (30 min)		(Phenrat <i>et al.</i> , 2016)
	Nanohybrid	nZVI-rGO	Oxidation	~100% (30 min)		(Gu <i>et al.</i> , 2018)
	Nanohybrid	Me-GO (Me = Ag, Co, Pt)	Adsorption		171.8–365	(Nas <i>et al.</i> , 2019;Rohaizad <i>et al.</i> , 2020)
	Nanohybrid	Fe-CNTs	Redox, filtration	97%		(Yanez <i>et al.</i> , 2017)
Nanohybrid	polyethylenimine surface-modified nZVI (PEI-nZVI)	Reduction	99% (120 min)		(Lin <i>et al.</i> , 2018)	
Tetrachloroethylene (PCE)	Nanocomposite	Fe-biochar	Redox	98.5% (420 min)		(Meng <i>et al.</i> , 2021)
	Nanocomposite	polyvinyl alcohol coated calcium peroxide nanoparticles/FeS (PVA@nCaO ₂ /FeS)	Redox	58.9%–99.7 (360 min)		(Ali <i>et al.</i> , 2020)
	Nanocomposite	polyethylenimine surface-modified nZVI (PEI-nZVI)	Reduction	99% (120 min)		(Lin <i>et al.</i> , 2018)
Nanocomposite	nZVI-layered double hydroxide (LDH)	Reduction & adsorption	100% (4 days)		(Wang <i>et al.</i> , 2020b)	
<i>Non-chlorinated compounds</i>						
Naphthalene	Carbonaceous	Multiwall carbon nanotubes (MWCNT-OH)	Adsorption		0.38	(Akinpelu <i>et al.</i> , 2019)
	Nanohybrid	ZnO/Ag/GO	Photodegradation	92% (50 min)	500	(Mukwevho <i>et al.</i> , 2020)
	Nanohybrid	Fe-ZnO nanofiber	Photodegradation	96% (240 min)		(Sekar <i>et al.</i> , 2018)
	Nanocomposite	Mag-PCMA	Adsorption	95% (200 min)		(Wang <i>et al.</i> , 2009)
	Nanocomposite	FeMn/biochar/H ₂ O ₂	Photodegradation	82.2% (148 min)		(Li <i>et al.</i> , 2019b)
Methyl orange	Nanocomposite	Kaolin/ Fe ₃ O ₄	Adsorption	97% (66 min)		(Arizavi <i>et al.</i> , 2019)
	Carbonaceous	MWCNTs	Adsorption		106.3	(Ahlawat <i>et al.</i> , 2020)
	Carbonaceous	MWCNTs/activated carbon	Adsorption		196.1	(Ahlawat <i>et al.</i> , 2020)
	Me/MeO	Co ₃ O ₄	Adsorption		46.082	(Uddin and Baig, 2019)
	Me/MeO	MgO ₂	Adsorption	97.4% (50 min)	2.01 (10 min)	(El-Shamy, 2020)
	Me/MeO	TiO ₂	Photodegradation	45.9% (UV) 26.7% (sunlight), (45 min)		(Pirsaheb <i>et al.</i> , 2020)

Table 3 (Continued)

<i>Contaminant</i>	<i>Technology</i>	<i>Method</i>	<i>Mechanisms</i>	<i>Efficiency</i>	<i>Capacity (mg/g)</i>	<i>References</i>
Methylene blue	Me/MeO	Fe ₃ O ₄	Oxidation	43% (30 min)		(Arshad <i>et al.</i> , 2018)
	Me/MeO	L-arginine-coated Fe ₃ O ₄	Adsorption		338.98	(Guo <i>et al.</i> , 2018b)
	Nanohybrid	Scoria-Ni/TiO ₂	Photodegradation	95.89% (UV), 93.97% (sunlight) (45 min)		(Pirsaheb <i>et al.</i> , 2020)
	Nanohybrid	SiO ₂ -Al ₂ O ₃ @Ni-H ₃ PW ₁₂ O ₄₀	Photodegradation	86% (180 min)		(Shalaby <i>et al.</i> , 2018)
	Nanohybrid	Graphene/Fe ₃ O ₄	Redox (oxidation)	99.24% (30 min)		(Arshad <i>et al.</i> , 2018)
	Nanohybrid	UiO-66-NH ₂ MOF	Filtration	84.5%		(Hashem <i>et al.</i> , 2019)
	Carbonaceous	MWCNTs	Adsorption		185.1	(Ahlawat <i>et al.</i> , 2020)
	Me/MeO	CuO	Adsorption	98.89% (210 min)	95.5	(Thakur and Kumar, 2019)
	Nanohybrid	GO/nZVI	Adsorption	99.1% (5 min)		(Mehrabi <i>et al.</i> , 2019)
	Nanohybrid	Pt-Co@GO	Adsorption		273.6	(Nas <i>et al.</i> , 2019)
	Nanohybrid	Ni/Zn-MOF-5	Photodegradation, adsorption	90% (100 min)		(Zebardast <i>et al.</i> , 2018)
	Nanohybrid	Co/Zn-MOF-5	Photodegradation, adsorption	90% (100 min)		(Zebardast <i>et al.</i> , 2018)
	Nanocomposite	Fe ₃ O ₄ -Si-COOH (silica gel)	Adsorption		246.31	(Guo <i>et al.</i> , 2018a)
Nanocomposite	MWCNTs/activated carbon	Adsorption		232.5	(Ahlawat <i>et al.</i> , 2020)	
Nanocomposite	ZnO-activated carbon	Adsorption	> 99% (60 min)		(Kamaraj <i>et al.</i> , 2020)	

Table 4 Recent studies about applying nanomaterials for disinfection byproducts removal

Contaminant	Technology	Method	Mechanisms	Efficiency	Capacity (mg/g)	References
<i>Disinfection byproducts</i>						
Trichloromethane (TCM)	Carbonaceous	MWCNTs	Adsorption		10.98	(Dehghani <i>et al.</i> , 2016)
	Carbonaceous	GO	Adsorption & photodegradation	70.6% (dark) 98% (visible light) (30 min)		(Ulucan-Altuntas <i>et al.</i> , 2020)
Bromodichloromethane (BDCM)	Me/MeO	Fe _{3-x} Co _{x4}	Redox	90% (5 min)		(Silva <i>et al.</i> , 2019)
	Carbonaceous	MWCNTs	Adsorption		6.85	(Dehghani <i>et al.</i> , 2016)
Dibromochloromethane (DBCM)	Me/MeO	Fe _{3-x} Co _{x4}	Redox	90% (5 min)		(Silva <i>et al.</i> , 2019)
	Carbonaceous	MWCNTs	Adsorption		6.57	(Dehghani <i>et al.</i> , 2016)
Tribromomethane (TBM)	Me/MeO	Fe _{3-x} Co _{x4}	Redox	87% (5 min)		(Silva <i>et al.</i> , 2019)
	Carbonaceous	MWCNTs	Adsorption		5.95	(Dehghani <i>et al.</i> , 2016)
Trichloronitromethane	Me/MeO	Fe _{3-x} Co _{x4}	Redox (oxidation)	86% (5 min)		(Silva <i>et al.</i> , 2019)
	Nanohybrid	Graphene-nZVI	Adsorption & redox	99% (120 min)		(Chen <i>et al.</i> , 2016)
Monochloroacetic acid (MCAA)	Carbonaceous	GO	Adsorption	31.6% (2 min)		(Liu <i>et al.</i> , 2020c)
Dibromoacetic acid (DCAA)	Carbonaceous	GO	Adsorption	27.1% (2 min)		(Liu <i>et al.</i> , 2020c)
	Me/MeO	Ti ₄ O ₇	Reduction	73% (11 s)		(Almassi <i>et al.</i> , 2020)
	Me/MeO	CuO+UV	Photodegradation	99.79% (75 min)		(Alavi <i>et al.</i> , 2021)
	Nanohybrid	MWCNT-Ti ₄ O ₇	Reduction	96% (11 s)		(Almassi <i>et al.</i> , 2020)
Trichloroacetic acid (TCAA)	Nanocomposite	Powder activated carbon-Ti ₄ O ₇	Reduction	94% (11 s)		(Almassi <i>et al.</i> , 2020)
	Carbonaceous	GO	Adsorption	30.2% (2 min)		(Liu <i>et al.</i> , 2020c)
	Me/MeO	CuO+UV	Photodegradation	99.22% (75 min)		(Alavi <i>et al.</i> , 2021)

Per- and polyfluoroalkyl substances (PFAS)

Use of carbonaceous nanomaterials (C-ENMs)

Research on the removal of PFAS by C-ENMs include adsorption, and redox reaction (see Fig. 1(c) and examples in Table 1) (Xue *et al.*, 2015; Elanchezhyan *et al.*, 2020; Masud *et al.*, 2021; Du *et al.*, 2014; Zhang *et al.*, 2019c). The high electron transport efficiency in C-ENMs enables them to act as precursors in redox reactions (Masud *et al.*, 2021; Li *et al.*, 2017). A recent study applied electrochemical oxidation process with CNTs to degrade perfluorooctanoic acid (PFOA), with a removal efficiency of 90% in 3 h (Xue *et al.*, 2015). Due to the hydrophobicity of most PFASs, adsorption based on hydrophobic interactions is another possible mechanism of PFAS removal by C-ENMs (Du *et al.*, 2014; Zhang *et al.*, 2019d).

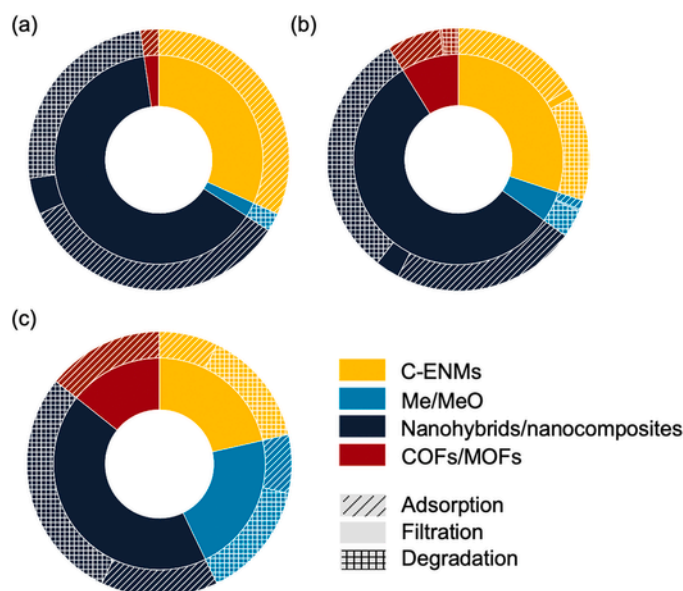


Fig. 1 Studies considering different nanomaterials to remove emerging contaminants from water based on different mechanisms. (a) Microplastics; (b) Pharmaceuticals and Personal Care Products (PPCPs) and Endocrine Disrupting Compounds (EDCs); and (c) Per- and Polyfluoroalkyl Substances (PFASs). The inner circle refers to four classes of nanomaterials (with different colors). The outer circle displays the mechanisms applied for treatment (with different textures). The definitions of acronyms in the figure: C-ENMs: carbonaceous-engineered nanomaterials; Me/MeO: nano metal/metal oxides; COFs: covalent organic frameworks; MOFs: metal-organic frameworks. The data was collected from the Web of Science considering the last 5 years (2016–2021).

Use of metal and metal oxides (Me/MeO)

There are quite a few studies related to adsorbing PFASs with nano Me/MeO (Clark *et al.*, 2019; Park *et al.*, 2018). For example, the adsorption capacity of UiO-66 for perfluorooctane sulfonate (PFOS) was 1.24 mg/g (Clark *et al.*, 2019). Compared with adsorption and filtration, degradation of PFAS appears more promising since it can destroy the chemicals and actually remove them from the water cycle. Me/MeO nanomaterials with high electron transport efficiency and reactivity have been widely studied for PFAS degradation (Chen *et al.*, 2015; de S. Furtado *et al.*, 2021; Xu *et al.*, 2017b; Li *et al.*, 2016). NanoTiO₂ is a very promising candidate for degrading PFAS by photocatalysis. Doping with high conductive metals, such as gold (Au), Ag, and copper (Cu), can improve the removal of PFAS (Chen *et al.*, 2015; Li *et al.*, 2016). For example, Cu-nanoTiO₂ photodegraded 91% of PFOA within 12 h (Chen *et al.*, 2015). In addition to photodegradation, PFOS can be oxidized and degraded by mixing MnO₂ and H₂O₂, and about 99% of pollutants can be removed in only 15 min (Chang *et al.*, 2020).

Use of nanohybrids/nanocomposites

There is considerable interest in employing nanohybrid and nanocomposite materials for removing PFAS (see Table 1), specifically designing the nanomaterials to improve degradation reaction efficiency (Fig. 1(c)) (Masud *et al.*, 2021; Zhao *et al.*, 2013; Li *et al.*, 2013; Huang *et al.*, 2016c; Jiang *et al.*, 2016). For example, Pb-BFeO₃/rGO can remove 90% of PFOA through Fenton reaction within 5 min (Li *et al.*, 2017). nZVI-rGO can remove 84% of PFOS within 10 min (Masud *et al.*, 2021). Also, nickel (Ni)-doped nZVI loaded on activated carbon was used to defluorinate PFOS (Zenobio *et al.*, 2020). About 94% of PFOS was decomposed in water at 50°C (Zenobio *et al.*, 2020). The doping of nZVI with the Ni catalyst enhanced the degradation PFOS, rather than mere adsorption without the catalyst (Zenobio *et al.*, 2020). Some nanohybrid materials have also been used in adsorbing PFAS (Elanchezhyan *et al.*, 2020; Omo-Okoro *et al.*, 2021). For example, Ag-activated carbon materials exhibited an adsorption efficiency of more than 80% for 10 different PFASs, and the adsorption capacity for PFOS (454.1 mg/g) was much higher than using traditional particulate activated carbon (PAC) technology (0.64 mg/g) (Clark *et al.*, 2019; Omo-Okoro *et al.*, 2021). Also, a Pb/nZVI hybrid had good PFOS removal efficiency (~35%) under acidic conditions (pH = 3.6) and 70°C. Complexation between PFOS and the dissolved Fe (III) from nanoparticles led to a better remediation performance (Park *et al.*, 2018).

Use of organic framework nanostructures

Although there are only a few studies on the use of emerging nanostructured materials for removing PFAS, the high surface area and porosity of COFs and MOFs are amenable for removing PFAS from water via adsorption (Fig. 1(c)) (Ji *et al.*, 2018; Guo *et al.*, 2020a). For example, [NH₂]-COFs achieved an adsorption efficiency of more than 90% for 13 PFAS compounds within 0.5 h (Table 1), which is very promising (Ji *et al.*, 2018). Additionally, a study found that the adsorption capacity of PFOS by organic ligand (biphenyl-4,4'-

dicarboxylic acid, BPDC)-modified MOFs could reach 304.9 mg/g, which was significantly higher than unmodified MOFs (66.2 mg/g) and the traditional PAC adsorption (0.64 mg/g) (Clark *et al.*, 2019; Zhao *et al.*, 2021).

Inorganic Chemicals

Metals

Use of carbonaceous nanomaterials (C-ENMs)

Given their high efficiency as adsorbents, C-ENMs have been widely used for removing metals in laboratory-scale studies (see Fig. 2(a) and some examples in Table 2). CNTs and GO can effectively remove metals such as Cu, (Dou *et al.*, 2019; Safari *et al.*, 2019; Bai *et al.*, 2019) Cd, (Vajedi and Dehghani, 2019; Salawudeen *et al.*, 2020) Cr, (Samuel *et al.*, 2019) and Pb (Vajedi and Dehghani, 2019) in water. The adsorption capacities of C-ENMs for heavy metals without modification are usually about 5–50 mg/g (see Table 2). Further modification of carbon nanomaterials can improve the remediation performance by increasing their adsorption capacity or via other mechanisms. For example, chitosan-coated CNTs absorbed Cu up to 115.5 mg/g, which is significantly higher than using the pristine CNTs (16.6 mg/g) (Dou *et al.*, 2019; Safari *et al.*, 2019). The increased capacity for Cu removal was due to the abundant amino and hydroxyl groups present in chitosan. In addition, modified carbon nanorods with 2,2'-(ethylenedioxy)bis(ethylamine) (EDEA) could adsorb and reduce Cr (VI), and 99% removal efficiency was achieved within 3 h (Anand *et al.*, 2019).

Use of metal and metal oxides (Me/MeO)

Removal of dissolved metals by Me/MeO using adsorption and redox reaction has been reported (see Fig. 2(a)). The toxicity, and thus, health risk of Cr (VI) can be decreased by reducing it to a lower valent state, Cr (III) (Barrera-Díaz *et al.*, 2012; Jin *et al.*, 2016). Iron-based nanomaterials, such as nZVI, have been applied for Cr (VI) reduction (Černíková *et al.*, 2020; Němeček *et al.*, 2016). Similar results can be achieved by using nano boehmite (γ -AlO), which had a adsorption capacity of 64.7 mg/g for Cr (VI) (Li *et al.*, 2019c). nZVI can also adsorb metals such as Cd, Ni, molybdenum (Mo), cobalt (Co), and Zn (see Table 2) (Su *et al.*, 2014a, 2015, 2018b, 2020, 2016). Sulfidation of nZVI and different ratios of sulfur to iron (S/Fe ratio) can impact metal removal due to the possibility of co-precipitation or ion exchange with FeS within SnZVI (Su *et al.*, 2015, 2020, 2016).

Use of nanohybrids/nanocomposites

Since nanohybrids/nanocomposites combine the characteristics of several materials, the mechanisms used for heavy metal removal include adsorption, filtration, and reactions (see Fig. 2(a), and some latest examples were listed in Table 2). After doping cobalt sulfide (CoS) on SWCNT, the adsorption capacity of the nanohybrid for mercury (Hg) was as high as 1660 mg/g (Alijani and Shariatinia, 2018). A nano-MoS₂@fiber composite had an adsorption capacity of 6258.7 mg/g for Hg (Fausey *et al.*, 2020). Nanocomposite materials have also been considered for filtration of metals. A polyetherimide based polyhedral oligomeric silsesquioxane (POSS) nanofiltration membrane filtered 85% of Cu and 86% of Pd (Bandeali *et al.*, 2019). In addition, many recent studies also explored nanohybrid/nanocomposite materials for enhanced redox-based treatment (Mohagheghian *et al.*, 2018; Nasiri *et al.*, 2018; Zhao *et al.*, 2019). For instance, WO₃-Fe₃O₄ nanocomposite could remove 99.96% of Cr (VI) in 5 h through photocatalytic reduction under visible light. The addition of Fe₃O₄ enhanced the electron transport efficiency and made it easy to recover the nanocomposite via external magnetic field (Mohagheghian *et al.*, 2018). Compared with traditional photocatalytic technology, the improved photodegradation performance of the nanocomposite was due to a smaller energy band gap, resulting in the lower light frequency required to excite electrons.

Additionally, ligand-functionalized nanoparticle sorbents (such as Mag-Ligand, which is functionalized with ethylenediaminetetraacetic acid (EDTA)) is efficient for removing several metals such as Pb, Hg, and Cd (Huang and Keller, 2015; Huang and Keller, 2016). The complexation reaction between the metals and the ligand on the particle surface enhanced the adsorption capacity (Huang and Keller, 2016).

Use of organic framework nanostructures

Several studies have explored the ability of MOFs and COFs to remove heavy metals, mostly based on adsorption (see Fig. 2(a) and some examples in Table 2). An amide-based COF had an adsorption capacity of 185.7 mg/g for Pd (II) (Li *et al.*, 2019d). The adsorption

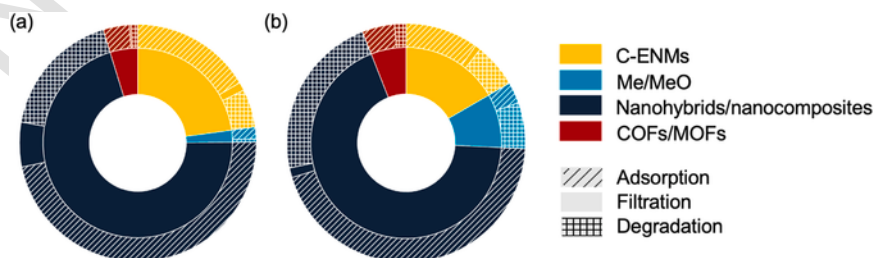


Fig. 2 Studies considering different nanomaterials to remove inorganic contaminants from water based on different mechanisms. (a) metals; (b) non-metals. The inner circle refers to four classes of nanomaterials (with different colors). The outer circle displays the mechanisms applied for treatment (with different textures). The data was collected from the Web of Science considering the last 5 years (2016–2021).

capacity of dual-porous COFs for Cr (VI) was as high as 384 mg/g, (Cui *et al.*, 2019) which is higher than the adsorption capacity of most other adsorbents for Cr (VI). Similarly, efficient removal of Cd (116 mg/g) by COFs (Liu *et al.*, 2020b) and Cr (225–259.1 mg/g) by MOFs (Fang *et al.*, 2020; Niu *et al.*, 2021; Zhou *et al.*, 2021) were also reported. These studies demonstrate that COF/MOFs exhibited better adsorption capacities than traditional GAC adsorption technology (the adsorption capacities for Pb (II), Cr (VI), and Cd (II) were 13.3 mg/g, 2.8 mg/g, and 10.3 mg/g, respectively) (Sulaymon *et al.*, 2009; Hydari *et al.*, 2012).

Metalloids and other non-metallic contaminants

Use of carbonaceous nanomaterials (C-ENMs)

Several studies have demonstrated that C-ENMs have good adsorption capacity for metalloids and other non-metal contaminants. A recent study an adsorption capacity of SWCNTs to be 49.65 mg/g for As (Ma *et al.*, 2018). In addition, several studies also explored the use of C-ENMs to remove nitrate (Table 2) (Ahmadzadeh Tofighy and Mohammadi, 2012; Alimohammadi *et al.*, 2016). The functionalization of oxidized-CNT with nitrogen-containing groups increased nitrate adsorption (from 90.9 mg/g to 142.9 mg/g) by enhancing the basicity of the material surface and diffusion driving force (Ahmadzadeh Tofighy and Mohammadi, 2012).

Use of metal and metal oxides (Me/MeO)

Me/MeO remove non-metal contaminants via adsorption and reduction (see Fig. 2(b) and Table 2). Some iron-based nanomaterials, such as nZVI and Fe₂O₃, have been reported to efficiently remove As, nitrate, and phosphate (Wen *et al.*, 2014; Han *et al.*, 2021; Wei *et al.*, 2018). Other Me/MeO nanomaterials, such as nanoCuO, nano γ -AlO, were also observed to possess high adsorption capacities for nitrate (Table 2) (Rahdar *et al.*, 2021; Li *et al.*, 2019c). Although nZVI is able to remove nitrate via reduction, a study found that the Pb adsorption by nZVI could be affected when nitrate is also present (Su *et al.*, 2014b). Initially, the formation of ferrite during nitrate reduction led to higher Pb adsorption. However, the excess of nitrate could further consume ferrite and cause the desorption of Pb from nZVI.

Use of nanohybrids/nanocomposites

Several studies have explored the use of C-ENMs combined with nanosized Me/MeO (nanohybrids), or nanocomposites of Me/MeO nanomaterials and activated carbon or polymers to remove non-metallic contaminants (Table 2). The main mechanism of removal of non-metals by C-ENMs is adsorption, but when combined with other materials, additional mechanisms, such as reduction, may become dominant (Fig. 2(b)) (Han *et al.*, 2020a; Motamedi *et al.*, 2014). For example, a study revealed that the introduction of GO onto the nZVI's oxide shell increased nitrate removal efficiency from 40% (when treated with nZVI only) to 80% within 0.5 h by improving the adsorption ability and electron transport efficiency (Han *et al.*, 2020a). Another study found that the removal rate of nitrate by nZVI increased from 32.8% to 78.2% after doping with biochar (Wei *et al.*, 2018). Also, a nanocomposite made up of chitosan and nano-biopolymer adsorbed 100% of nitrate (Ghaemi *et al.*, 2018).

Mag-PCMA was effective for the adsorption of several anions (including perchlorate, phosphate, nitrate, and sulfate) due to the abundance of cationic sites on the particle surface (Clark and Keller, 2012b). Nanomaterials combined with polymers are also promising for filtration of non-metals. For instance, CeO₂ and ceramic fiber nanocomposite achieved 94.3% removal of As due to both adsorption and filtration (Yang *et al.*, 2021).

Use of organic framework nanostructures

Adsorption is the most common mechanism for COFs and MOFs to remove non-metallic pollutants (see Fig. 2(b) with some studies summarized in Table 2). A recent study determined the adsorption capacity of Zr-MOFs for As (III) and As (V) was 211 and 325 mg/g, respectively, (Yu *et al.*, 2019) which are higher than that of GAC adsorption technology (38.8 mg/g and 51.3 mg/g for As (III) and As (V), respectively) (Chen *et al.*, 2007). In addition, COFs modified with ethidium bromide was also efficient for As removal, with an adsorption capacity of 53.1 mg/g (Yang *et al.*, 2020). A recent study indicated that the adsorption and chelation between Fe-MOFs and nitrate resulted in a high adsorption capacity of 92.6 mg/g (Pandi and Choi, 2021).

Organic Compounds

Halogenated organics

Use of carbonaceous nanomaterials (C-ENMs)

Several studies have applied C-ENMs for adsorption of halogenated organics (Fig. 3(a)), and high adsorption capacity was reported for trichloroethylene (TCE) and tetrachloroethylene (PCE) (Table 3). In a recent study, CNTs was used to filter TCE in water, with a removal rate of up to 74% (Yanez *et al.*, 2017). Another study investigated the interaction energy (IE) between chlorobenzenes and two C-ENMs (SWCNT and graphene) based on density functional theory (DFT) and electronic structure calculations. The authors found that the IE was higher with higher number of chlorine atoms in the contaminants, which suggests that adsorption capacity would increase with chlorination of the organic compounds (Balamurugan and Subramanian, 2013).

Use of metal and metal oxides (Me/MeO)

Some Me/MeO nanomaterials are likely used to degrade halogenated organics (Fig. 3(a)), due to the nanomaterials' high electron transport efficiency and low energy band gap. Among others, nanoTiO₂ and nZVI have been widely employed to degrade halogenated organic compounds (Meng *et al.*, 2021; Phenrat *et al.*, 2016; Shiraishi *et al.*, 2011). nZVI has a high reduction potential, but the efficiency of TCE degradation was very limited (3.38% in 420 min) (Meng *et al.*, 2021). Another study indicated that sulfidation of nZVI

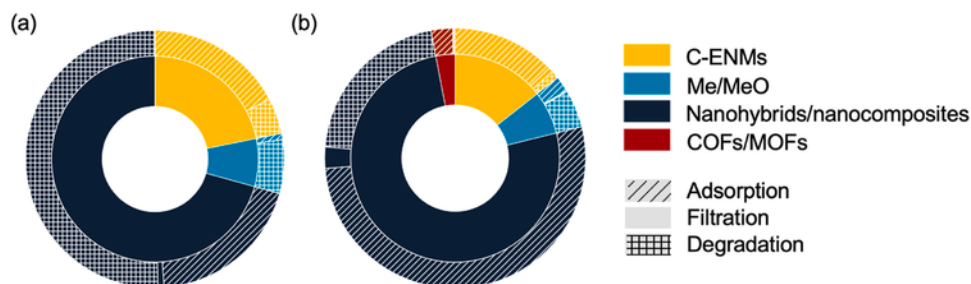


Fig. 3 Studies considering different nanomaterials to remove organic contaminants from water based on different mechanisms. (a) halogenated organics (b) non-halogenated organics. The inner circle refers to 4 types of nanomaterials (with different colors). The outer circle displays the mechanisms applied for treatment (with different textures). The data was collected from the Web of Science considering the last 5 years (2016–2021).

significantly increased the surface area-normalized degradation rate constant of TCE to around 50-fold higher than pristine nZVI, by improving electron transport efficiency (Xu *et al.*, 2020). In addition, a recent study found that applying nZVI under an alternating current electromagnetic field can reduce 100% of TCE within 30 min (Phenrat *et al.*, 2016). Other studies that used Me/MeO for halogenated organics removal were listed in Table 3.

Use of nanohybrids/nanocomposites

Nanohybrids/nanocomposites are mainly used to improve the adsorption or degradation efficiency of halogenated organics by a single nanomaterial (Fig. 3(a)). Given the relatively weak performance of nZVI for TCE, (Meng *et al.*, 2021) nanocomposites have been developed with different modification approaches, such as doping nZVI with rGO, double-layer hydroxyl (LDH), or polyethyleneimine (PEI), respectively. The nanocomposites have improved reactivity and TCE removal efficiency (more details were shown in Table 3) (Lin *et al.*, 2018; Wang *et al.*, 2020b; Gu *et al.*, 2018). For instance, depositing nZVI onto CNTs membranes increased adsorption and reduction of TCE, and ultimately increased removal efficiency from 74% (for CNT membranes) to 97% (for Fe-CNT membranes) (Yanez *et al.*, 2017).

Non-halogenated organics

Use of carbonaceous nanomaterials (C-ENMs)

Existing studies have shown that C-ENMs are used for adsorbing non-halogenated organic compounds such as naphthalene, methyl orange, methylene blue, among others (Fig. 3(b)). The adsorption efficiency depends strongly on the contaminant (Table 3). For example, the adsorption capacity of MWCNT for naphthalene was only 0.38 mg/g, but it was 106.3 for methyl orange and 185.1 mg/g for methylene (Akinpelu *et al.*, 2019; Ahlawat *et al.*, 2020). Furthermore, a composite of MWCNT and activated carbon had an even higher adsorption capacity for methyl orange and methylene—196.1 and 232.5 mg/g, respectively (Ahlawat *et al.*, 2020). Hydrophobic interactions play an important role in the adsorption of organic compounds by C-ENMs. As an example, the exposure of GO to light increased its ability to adsorb phenanthrene due to increased hydrophobicity of GO upon light exposure (Du *et al.*, 2018).

Use of metal and metal oxides (Me/MeO)

The mechanisms for removing non-halogenated organics using Me/MeO nanomaterials mainly include adsorption and redox reactions (see Fig. 3(b) and some recent examples in Table 3). NanoTiO₂ is the most popular nanomaterial used for photocatalytic degradation (Pirsaheb *et al.*, 2020; Rashed and El-Amin, 2007; Yogi *et al.*, 2008). A recent study found that the removal efficiency of methyl orange (45.9%) by commercial nanoTiO₂ under UV light is higher than that under sunlight (26.7%) (Pirsaheb *et al.*, 2020). Also, nanoCo₃O₄, nanoMgO₂, and nanoCuO have been used as adsorbents for dyes such as methyl orange and methylene blue (Uddin and Baig, 2019; El-Shamy, 2020; Thakur and Kumar, 2019). Some iron-based nanomaterials, such as nanoFe₃O₄, can also remove methyl orange from water through redox reaction (Arshad *et al.*, 2018).

Use of nanohybrids/nanocomposites

Nanohybrid/nanocomposite are commonly used to achieve improved adsorption and redox efficiency for non-halogenated organics (see Fig. 3(b) and some recent examples in Table 3). Hybridizing GO with metals (such as Ag, Co, and Pt) enhanced the GO's methylene blue adsorption capacity from 137.5 mg/g to 171.75–365.0 mg/g for the metal-GO nanohybrids (Nas *et al.*, 2019; Rohaizad *et al.*, 2020). In addition, hybridization or compositing of some nanomaterials can also improve electron transport efficiency. For example, a scoria-Ni/TiO₂ nanocomposite photodegraded >90% of methyl orange under UV and sunlight, which was much higher than the using nanoTiO₂ alone (Pirsaheb *et al.*, 2020). Another study found that the depositing nanoFe₃O₄ onto graphene increased the removal efficiency of methyl orange from 43% (for only nanoFe₃O₄) to 99%, and the spent nanohybrid could be easily removed with a magnetic field (Arshad *et al.*, 2018).

Additionally, Mag-PCMA has affinity for several polycyclic aromatic hydrocarbons (PAHs) (e.g., acenaphthene, naphthalene, and biphenyl) due to the hydrophobic surfactant coating (Wang *et al.*, 2009; Clark and Keller, 2012a). The presence of humic acid had a

limited effect on the adsorption of PAHs, indicating a good performance when treating water with high content of NOM (Wang *et al.*, 2009). Also, the lower costs of Mag-PCMA compared to current treatment technology make it promising in future applications (Adeleye *et al.*, 2016a).

Use of organic framework nanostructures

Several recent studies on organic framework nanostructure are related to adsorption by metal-doped MOFs (see Fig. 3(b) and Table 3). Among them, a UiO-66-NH₂ modified MOF was applied for adsorbing methyl orange from wastewater, and the efficiency could reach 84.5% (Hashem *et al.*, 2019). In addition, MOFs modified with Ni, Co, and Zn could simultaneously adsorb and photodegrade methyl blue, with a removal efficiency of 90% within 100 min (Zebardast *et al.*, 2018). Although there are fewer studies on the use of COFs and MOFs for non-halogenated compounds, their high porosity and ease of modification make them promising materials for adsorption of non-halogenated organic compounds.

Disinfection Byproducts

Use of carbonaceous nanomaterials (C-ENMs)

The application of C-ENMs in the removal of disinfection by-products (DBPs) is mainly based on adsorption (see Fig. 4 and Table 4) (Dehghani *et al.*, 2016; Liu *et al.*, 2020c; Silva *et al.*, 2019). The adsorption capacity of unfunctionalized MWCNTs for four trihalomethanes (THMs), trichloromethane (TCM), dibromochloromethane (DBCM), bromodichloromethane (BDCM), and tribromomethane (TBM), was reported as 10.98 mg/g, 6.85 mg/g, 6.57 mg/g, and 5.95 mg/g, respectively (Dehghani *et al.*, 2016). The higher capacity for TCM compared to other THMs was due to the higher polarity and lower molecular weight of TCM, which led to higher penetration into MWCNTs (Dehghani *et al.*, 2016). On another note, GO was reported to rapidly adsorb 31.6% of monochloroacetic acid (MCAA), 27.1% of dibromoacetic acid (DCAA), and 30.2% of trichloroacetic acid (TCAA) within 2 min, respectively. The mechanism of adsorption was mainly via formation of hydrogen bonds between GO's functional groups and the contaminants (Liu *et al.*, 2020c).

Use of metal and metal oxides (Me/MeO)

Metal nanomaterials are another type of nanomaterials commonly considered for removing DBPs. In contrast with C-ENMs, current research is more focused on the degradation of DBPs (see Fig. 4 and Table 4). For example, nanoFe_{3-x}Co_x4 with H₂O₂ degraded more than 85% of TCM, DBCM, BDCM, and TBM in 5 min through the Fenton reaction (Silva *et al.*, 2019). The degradation pathway was faster and more effective than adsorption with MWCNTs (Dehghani *et al.*, 2016). nZVI was also efficient for THMs and chloroform (CF) dichlorination because of its strong reducing ability (Xiao *et al.*, 2014; Feng *et al.*, 2008; Xiao *et al.*, 2015).

Use of nanohybrids/nanocomposites

Studies have reported the application of nanohybrids/nanocomposites in removing DBPs via adsorption and degradation (See Fig. 4 and Table 4). Among them, the reduction of dibromoacetic acid (DCAA) increased from 73% (when treated with nanoTi₄O₇ only) to 96% and 94% within 11 s after hybridizing nanoTi₄O₇ with MWCNT and hybridizing nanoTi₄O₇ with PAC, respectively (Almassi *et al.*, 2020). The addition of the adsorbents (MWCNTs and PAC) helped to concentrate the DBPs close to the catalyst, which improved the dehalogenation efficiency (Radjenović *et al.*, 2012; Fang and Al-Abed, 2007). Similar roles were found in THMs reduction by a nZVI/PAC composite, where the adsorption by activated carbon significantly increased the removal capacity of nZVI for THMs (Xiao *et al.*, 2015).

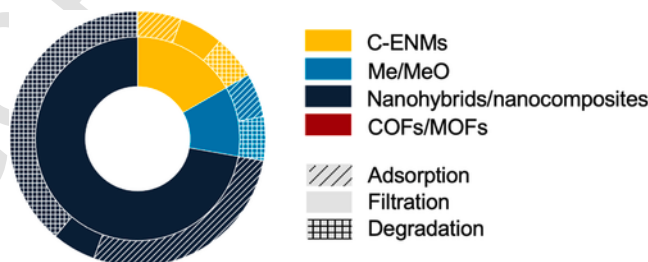


Fig. 4 Studies considering different nanomaterials to remove disinfection byproducts from water based on different mechanisms. The inner circle refers to 4 types of nanomaterials (with different colors). The outer circle displays the mechanisms applied for treatment (with different textures). The data was collected from the Web of Science considering the last 5 years (2016–2021).

Applications of Nanomaterials in Water Treatments

Commercial Considerations

Although the use of nanotechnology for water treatment is promising, some disadvantages have slowed progress towards commercial applications. For example, studies have demonstrated that nZVI has high adsorption capacity and reactivity for several contaminants (Babuponnusami and Muthukumar, 2012; Zha *et al.*, 2014; Mikhailov *et al.*, 2017; Adeleye *et al.*, 2013). However, nZVI reacts very rapidly once it is exposed to oxygen and/or oxygenated water, (Su *et al.*, 2020; Adeleye *et al.*, 2013, 2016b) releasing Fe^{2+} and losing its reduction potential, which affects its long-term remediation performance (Su *et al.*, 2015, 2018b). Several modifications of nZVI have been explored in response to these shortcomings, and the most popular among them is sulfidation. Sulfidation was found to slow down the dissolution and oxidation of zero-valent iron (Su *et al.*, 2015, 2020; Huang *et al.*, 2015). Many studies have found that the introduction of sulfhydryl groups improves the electron transport efficiency and redox capabilities, resulting in better removal efficiency for certain pollutants (Su *et al.*, 2015; Lv *et al.*, 2019; Cao *et al.*, 2017; Su *et al.*, 2020). Since SnZVI is also magnetic, it can be easily separated and recovered from water in real applications.

The cost of many nanomaterials, and the sustainability of the synthesis of many novel nanoparticles are also important considerations, if nanotechnology is going to succeed in the marketplace (Adeleye *et al.*, 2016a; Brame *et al.*, 2011; T C *et al.*, 2018). Nanotechnology also has to demonstrate reliability, ease of use, recoverability of the nanomaterials after they perform their water treatment function, reusability, and wherever possible recyclability (Qu *et al.*, 2013).

Environmental and Human Health Issues

Another factor that limits the wide application of nanomaterials in water treatment is the potential health and environmental impact of nanomaterials. Although some nanomaterials show high reactivity and could degrade contaminants, formation of byproducts or the reactivity of the nanomaterials themselves may have adverse effects on the environment or organisms (Stevenson *et al.*, 2017; Su *et al.*, 2018b; Brame *et al.*, 2011; Qu *et al.*, 2013; Garner *et al.*, 2015; Garner and Keller, 2014; Keller *et al.*, 2012; Cheng *et al.*, 2019a; Chen *et al.*, 2013; Saleh, 2020). Some nanomaterials with strong oxidation properties may damage cells or increase oxidative stress by producing reactive oxygen species (ROS) (Stevenson *et al.*, 2017; Cheng *et al.*, 2019a; Chen *et al.*, 2013; Saleh, 2020; Chen *et al.*, 2012). However, considerable efforts have made towards understanding the implications of and improving the sustainability of nano-enabled water treatment and nanoremediation (Han *et al.*, 2021; Cheng *et al.*, 2019a; Saleh, 2020; Thanh, 2013; Phenrat *et al.*, 2009). As an example, sulfidation decreases the toxicity of nZVI to several organisms (Han *et al.*, 2021; Cheng *et al.*, 2019a; Thanh, 2013) due to lower dissolution and oxidative stress (Keller *et al.*, 2012; Cheng *et al.*, 2019a; Chen *et al.*, 2013).

When nanomaterials are used as adsorbents, there is the risk that even a small amount of the nanomaterial loaded with contaminants can be discharged into the environment with treated water (Grassian, 2008; Zhang *et al.*, 2008; Weinberg *et al.*, 2011). Thus, any such application must guarantee full removal or immobilization of the nanoadsorbents. Similarly, reactive nanomaterials must be completely transformed and/or recovered, to avoid human or ecological exposure to these very active materials.

Combination With Conventional Technology

The combination of nanomaterials and traditional technology is another popular research direction in the application field. This approach can integrate the advantages of multiple treatment technologies and reduce the cost of redesigning and construction of new processes in water treatment. Nanomaterial incorporated with nanofiltration membranes is one of the approaches to improve the performance of filtration. Membrane combined with nano Me/MeO such as nanoTiO₂ and nano-Ag can improve the hydrophilicity of the membrane and anti-fouling properties (Andrade *et al.*, 2015; Mollahosseini and Rahimpour, 2014; Anand *et al.*, 2018). The C-ENMs-based membrane can enhance the separation performance of the membrane because of easily functionalized. For example, introducing amine-functionalized MWCNTs into the NF membrane could increase the hydrophilicity of the membrane and water permeation. And more functional groups resulted in higher surface charge and salt rejection (Zarrabi *et al.*, 2016).

Another approach is combining nanoTiO₂ with conventional AOP technologies (Jafarnejad, 2019; Suzuki *et al.*, 2015; Bethi *et al.*, 2016). Although traditional AOP technology has good removal efficiency for many residual contaminants, it requires a significant amount of energy and other expenses, limiting the application of this technology in water treatment plants (Adeleye *et al.*, 2016a; Jafarnejad, 2019; Mahamuni and Adewuyi, 2010). By introducing nanoTiO₂ into the AOP system, the increase rate of photodegradation can improve the efficiency of AOP and save energy required for the treatment (Suzuki *et al.*, 2015; Bethi *et al.*, 2016).

Additionally, some research explored the combination of nanomaterials with biological treatment (Yang *et al.*, 2019a; Almusawy *et al.*, 2020; Dixit *et al.*, 2011). CNTs were considered a promising material for membrane bioreactor (MBR) because of their high porosity and electron transport efficiency (Hinds *et al.*, 2004; Wei *et al.*, 2014). A recent study set up a CNTs-based electro-assisted MBR (EMBR) system to treat municipal wastewater (Yang *et al.*, 2019a). The effluent had less than 40 mg/L of chemical oxygen demand (COD) and 3 mg/L of $\text{NH}_4^+\text{-N}$ concentration with 4 h hydraulic retention time (HRT), which was significantly lower than that of two traditional PVDF-based MBR systems (over 50 mg/L of COD and 5 mg/L of $\text{NH}_4^+\text{-N}$ in effluent). Also, electrochemical oxidation could degrade the contaminants on the membrane surface and result in a better anti-fouling property.

Case Studies of Commercial Products Enabled With Nanomaterials

Combination of direct current electrode with nZVI to remove chlorinated ethenes (CEs) from groundwater

Due to its high reactivity, nZVI has become one of the most applied nanomaterials in commercial groundwater treatment. A recent study assessed the performance of commercial nZVI for removal of chlorinated ethenes (CEs) from groundwater (Černíková *et al.*, 2020). In the study, two different types of commercial nZVIs, TODA and NF 25S, were introduced into multiple wells, and regularly replenished to keep the nZVI concentration at 2–5 kg/m³. In the injected groundwater, the initial concentration of CEs was 8880 µg/L, dichloroethane (DCE) was the main component (about 7300 µg/L), while vinyl chloride (VC), perchloroethene (PCE), and trichloroethene (TCE) were present at lower amounts. After 25 months of nZVI loading, direct current (DC) electrodes were introduced into the reaction system to test the effect of an electric field on the remediation performance of nZVI. The electrodes were made of stainless steel (because of lower replacement cost), and the distance between the two electrodes was 3 m. The system had a 24 V DC, 7–15 A current, and the maximum output power to the well was 0.24 kW. The anode electrode was regularly replaced due to the adverse effects of aging and formation of oxide layer on electrode after long-term use performance.

Within the 64 months of monitoring, the researchers found that the concentrations of the CEs in groundwater decreased to 50–60% of the initial level after 4-month treatment with nZVI alone. However, due to the consumption of nZVI during the degradation of CEs and the production of byproducts, the concentrations of the pollutants rebounded to the original level or higher after 4 months. The two commercial nZVIs, which were from different manufacturers, had different removal efficiencies on CEs (~46% for TODA and ~60% for NF 25 S) after 4 months of remediation. When the electric field was introduced, the concentration of CEs decreased below 2000 µg/L (about 22% of the initial value) within 4 months. After 50 months and several regular anode replacements, CE concentrations further decreased to 210 µg/L (approximately 2% of the initial value). The concentration rebound was again observed when the electrode failure. The introduction of the electric field modified the degradation mechanisms since oxidation of chlorinated solvents at the anode produced mineralized products, and reduction of the chlorinated solvents at the cathode dechlorinated them into alkanes or a small amount of chlorinated intermediate products. Regarding the processing equipment cost, the study pointed out that when using nZVI alone, the system costs 28.7–34.4 EUR/g of CEs treated. However, introducing DC electrode decreased the cost to 5.2 EUR/g of CEs treated.

Combination of nZVI with biotechnology to treat Cr (VI) and chlorinated solvents in groundwater

In this study, nZVI was combined with biotechnology to remove Cr (VI) and chlorinated solvents in groundwater (Němeček *et al.*, 2016). The initial concentration of Cr (VI) in groundwater was 4.4–57 mg/L, and CEs were 400–6526 µg/L (where TCE and cis-DCE were the main species). Initially, nZVI was injected into the well to achieve an nZVI concentration of 1 g/L, and again 3 months later to reach a concentration of 2 g/L. Three months after the second nZVI injection, whey was injected and recirculation was started. In the next 10 months, whey was continuously supplemented to maintain the total organic compound (TOC) concentration in the groundwater at 60 mg/L.

When nZVI was initially injected, the concentration of Cr (VI) immediately decreased below the limit of quantitation (LOQ = 0.05 mg/L). However, around one month later, Cr (VI) concentration increased and eventually reached 10%–15% of the initial concentration. A similar decrease and rebound were also found during the second injection, after more nZVI was applied. A low removal efficiency of CEs was also observed during this period because of competition between CEs and Cr (VI). However, after adding whey the concentration of Cr (VI) and CEs decreased rapidly. The concentration of Cr (VI) was lower than the LOQ level, and the reduction product, Cr (III), was immobilized in the soil in oxide and hydroxide forms. CE concentrations after treatment were as low as 0.1–0.2 µg/L. The study indicated that the microorganisms in the whey reduced the oxidized iron (trivalent iron (Fe (III)) to divalent iron (Fe (II))). The Fe (II) acted as a reducing agent for Cr (VI) reduction and extended the service life of nZVI. Additionally, the toxicity of the groundwater was significantly reduced after treatment, indicating that the technology was effective. However, the Cr speciation in soil after the remediation and the possibility of the Cr remobilization in the future were not considered in the study.

Degradation of petroleum hydrocarbons in groundwater with nanoTiO₂ and AOP

Nanosized TiO₂ has a low cost, large surface area, high reactivity, and is considered a promising material for water treatment in the real environment. The small band gap in nanoTiO₂ leads to strong photoreactivity. NanoTiO₂ can generate hydroxyl radicals under solar light (especially UV light) and oxidize residual organic substances. Therefore, nanoTiO₂ is suitable for the AOP-based water purification. In a study, nanoTiO₂ was applied for the treatment of petroleum hydrocarbons in groundwater via sunlight irradiation (Cho *et al.*, 2006). Two different types of nanoTiO₂ (with a concentration of 1.0% wt.), nanoTiO₂ sludge and nanoTiO₂ immobilized on ceramic beads, were introduced into the photoreactor. The photoreactor unit comprised several quartz columns. The bottom and side of each column were surrounded by aluminum foil with high UV reflectivity. The entire reactor was perpendicular to the incident angle of the noon sun to obtain a high radiation area. The entire system consisted of 4 continuous reactors units (total radiation area was 3 m²), and the concentrations of benzene, toluene, ethylbenzene, and xylene (BTEX) and total petroleum hydrocarbons (TPH) in the effluent were monitored from noon to 4 pm. In addition, combinations of sunlight + H₂O₂ and sunlight + H₂O₂ + nanoTiO₂ were also used to test the treatment of BTEX and TPH.

Before the treatment, the initial BTEX and TPH concentrations in the groundwater were 46.9–103.0 mg/L and 584.0–954.0 mg/L, respectively. After 4 h of sunlight exposure, the removal of BTEX and TPH in 10 mM H₂O₂-treated group was only 40% and 10%, respectively, due to low generation of hydroxyl radicals from H₂O₂ under sunlight. Higher removal efficiencies were observed in the slurry nanoTiO₂-treated group (80% removal for BTEX and 76% for TPH) than immobilized nanoTiO₂-treated group (35% removal for BTEX and 11% for TPH) in the presence of sunlight. The difference in removal efficiency might be related to higher surface area of nanoTiO₂ in the slurry system than in the immobilized system. The addition of 10 mM H₂O₂ in sunlight + nanoTiO₂ system increased the

removal efficiency to 100% (BTEX) and 88% (TPH) in the slurry nanoTiO₂-treated group, and 66% (BTEX) and 50% (TPH) in the immobilized nanoTiO₂-treated group, indicating the promotion of photo-oxidation by H₂O₂.

Although a better remediation performance was found in the slurry nanoTiO₂-treated group, the recovery of slurry nanoparticles was difficult. Thus, the H₂O₂ + immobilized nanoTiO₂ is a better choice in real-world treatment. Another issue with this system is that the reactor may not work continuously due to the limited daylight time. Therefore, combining solar irradiation with an artificial UV light system may improve the overall efficiency of the system.

Conclusions and Recommendations

With the intensification of water resources issues, it will become more challenging for traditional water treatment technologies to meet increasingly stringent water quality standards, especially for emerging contaminants that most traditional technologies cannot remove efficiently. Although some existing advanced water treatment technologies can remove these hard-to-degrade pollutants, these technologies often consume a large amount of energy, and are not economical, especially when dealing with residual pollutants. There is considerable interest in nanomaterials for water treatment given their high adsorption and/or filtration capacities, and/or reactivity. Also, there is the potential for nanotechnology to be simple to operate and consume less energy. In addition, the performance of nanomaterials can be improved by hybridization or doping with other materials, making them applicable to different environmental conditions and pollutants. However, studies on using nanomaterials to remove DBPs and emerging contaminants (such as MPs and PFASs) are still lacking (Fig. 5). Thus, more research should consider the possibility of applying nanomaterials for removing these important persistent contaminants.

Nanomaterials have great potential in water treatment but many issues need to be further investigated and resolved. For example, the fate and transport, and toxic effects of residual nanomaterials after treatment must be fully assessed. Several studies have confirmed that some nanomaterials are toxic to organisms, making it imperative to effectively recover the nanomaterials after their use, or consider further modification to reduce their toxicity. In addition, most of the current studies are lab-scale or pilot-scale, and there is a need to consider how these novel materials can be incorporated into conventional treatment trains. Therefore, future studies should also consider the path to commercial application, including decreasing the cost of nanomaterial synthesis and processing.

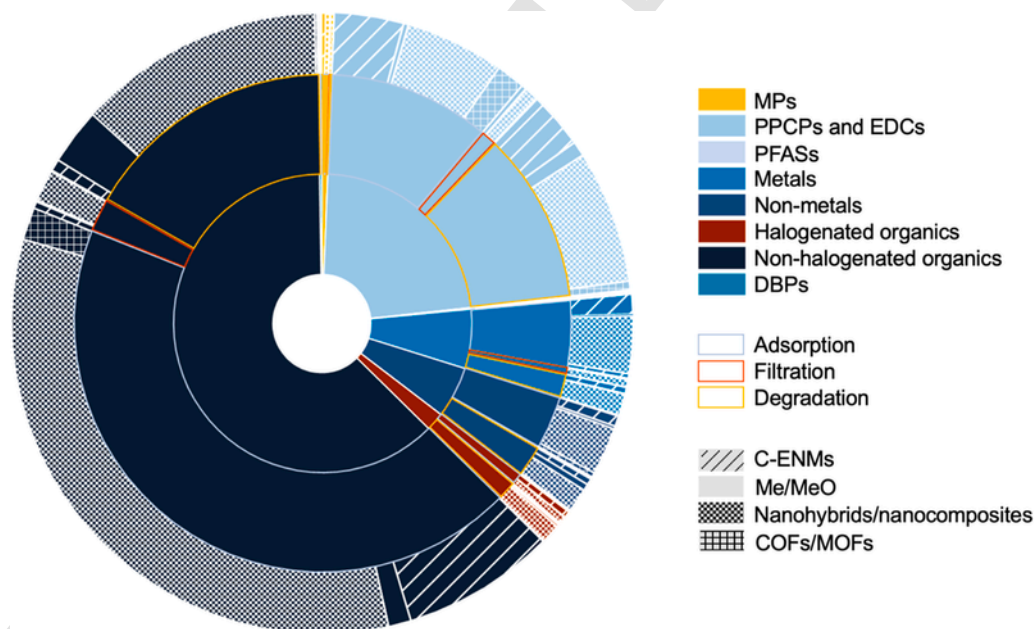


Fig. 5 Studies considering different nanomaterials to remove contaminants from water based on different mechanisms. The inner circle refers to 4 types of nanomaterials (with different colors). The middle circle displays the mechanisms applied for treatment (with different border colors). The outermost circle indicates the nanomaterials applied for treatment (with different textures). The data was collected from the Web of Science considering the last 5 years (2016–2021).

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